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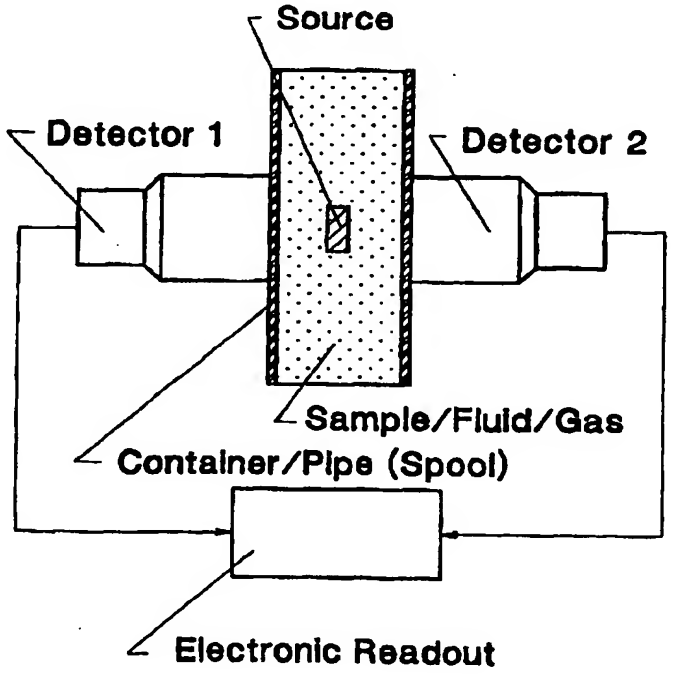
(54) Title: METHOD AND APPARATUS FOR REMOTE DENSITY MEASUREMENT

(57) Abstract

A Multiphoton Densitometer (MP-Densitometer) for remote density measurement uses special multiphoton sources with very low, licen se-exempt activity to measure the density of samples. For gas/oil/water mixture inside 3 inch steel pipes, the measurement precision, using a 2 microCi source, is substantially equivalent to that obtained by a prior-art gamma densitometer using a 30 milliCi source. Using a 1 microCi source the MP Densitometer measures the density, with uncertainty less than 1 %, of fluids inside pipes with a range of diameters up to 10 inches. The invention achieves very low background (orders of magnitude lower than for prior-art gamma densitometers) through selection of densitometer elements, geometry, electronics, and software. Reduction of system uncertainties, a temperature compensation scheme, active temperature regulation, real-time statistical analysis, and smoothing of the output signal allowed the elimination of temperature effects and pile-up artifacts. With a very low activity source, the MP-Densitometer has an accuracy in density measurements of 1 % for oil/water laminar flow. The uncertainty is 0.5 % for longer measurement tim es on the order of a few minutes.



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METHOD AND APPARATUS FOR REMOTE DENSITY MEASUREMENT

BACKGROUND OF INVENTION

The invention provides a method and apparatus for remote measurement of density of gas, fluids, solids or mixtures of the above. The disclosed method of MP-densitometry allows for a densitometer which is transportable, low cost and uses only low activity sources, *i.e.*, license exempt activity.

The invention relates to improvements in gamma densitometry using sources emitting several high energy photons simultaneously, and detecting the signals of these photons in coincidence to reduce the background hundreds of times and provide precision in density measurements better than that of the standard gamma densitometer but with a source with 10,000-fold lower activity than the one used in a standard gamma densitometry (license exempt activity). Such devices are referred to here as Multiphoton densitometers (MP-densitometers) or enhanced gamma densitometers (EGD).

In many applications, one needs to measure the density of a material in motion, either gas, liquid, solid, or granulated, *e.g.*, fluids and gases in pipelines or foils/flats/slabs of metal in the process of milling. Often, the material is either hazardous or can be easily contaminated. Examples of hazardous materials are those which are toxic, easily inflammable or explosive. For many biological and agriculture products spoilage or contamination results from contact with external air. However, the majority of classical methods of density measurement require sample taking or aliquoting and are not practical in disclosed applications.

Gamma densitometers are used to measure the parameters of bulk fluids. Especially important is the use of densitometers in characterization of petroleum and petroleum products, *e.g.*, measurement of relative fractions of petroleum, water and gas. Herein, the main problem is large variability of fluid composition and inhomogeneities. Statistical sampling of the properties is made very difficult by the presence of large bubbles of gas, thick layers of water and a combination of all types of small bubbles.

Gamma densitometers are also used for measurement of density of granulated material. For example, density is an important parameter when handling grains, hops and other agricultural products. Typically, it is easier to measure the density of grains in flow than

to measure their weight, grains size distribution, and then calculate the density. For example, the direct measurement of density permits estimation of the fraction of water in the sample.

Finally, there are applications when the material itself is solid; typically in form of a thin foil or plate. For example, in many metallurgic processes the foils and metallic flats of different thicknesses are produced by a milling from metal at very high temperature. The feedback between the parameters of milling machine and an instrument measuring the material thickness is required. Similarly gamma densitometers can be used in the paper industry to measure the thickness of paper or in chemical industries to measure the thickness of plastic foil. In all of these applications the gamma densitometer is actually used to measure thickness, because the density of solid materials is well known.

Densitometers are often used to measure properties of mixture(s) of fluids/gases/solids. The typical example is in the petroleum industry where crude oil is a mixture of petroleum, water and gas. In multiphase problem applications, the densitometers are used not only to measure the density of mixture, but are a part of more complex system that determines the composition of the crude oil. In this case, the dependence of absorption of high energy photons on the chemical composition can be used. Herein, a plurality of radiation sources with very different energies are used.

Many types of densitometers use the absorption of radiation (electromagnetic or sound waves) by matter. The ideal densitometer should have

* relatively low attenuation due to the container when compared with the attenuation in the sample;
* small variation of absorption coefficient with material composition;
* negligible effects of surface scattering.

In existing gamma densitometers, the attenuation of high energy photons, typically with energy larger than 300 keV, is measured in the configuration shown in comparative Figure 1. Photons emitted by the source are detected by a scintillator counter located on at the opposite side of the sample. Unfortunately, existing gamma densitometers have two limitations:

* high intensity sources are used, typically larger than 10 mCi;
* the density resolution is not better than a few percent.

These limitations of prior-art gamma densitometers can be traced back to:

* high radioactive background;

* a few percent drifts of the count rate due to temperature sensitivity of system components;
* instabilities, mainly due to pileups and dead-time artifacts.

SUMMARY OF THE INVENTION

The invention relates to a method for determining the density of a gas, liquid, solid or mixed sample comprising measuring the attenuation and preferably the scatter of at least one of a plurality of photons concurrently emitted by a radioisotope, wherein at least two of the photons are detected in coincidence in at least two detectors. The method may involve:

(a) placing a radioactive source capable of concurrently emitting at least two photons so that emitted photons pass through the sample,

(b) detecting at least two photons concurrently emitted by the radioisotope, in coincidence in at least two detectors, and

(c) measuring the attenuation in the sample of at least one of the photons concurrently emitted by the radioisotope.

The invention may employ long life positron-gamma radioemitters listed in Table 1, long life nuclear cascade radioemitters listed in Table 1, and long life electron capture radioemitters listed in Table 2. The total source activity may be below 10 microCi and background may be rejected in the photon detectors by (i) selecting low natural radioactive background elements; (ii) analyzing pulse height and pulse shape; or (iii) using coincidence signature, individually or in combination.

The method may further comprise shaping the signals from the detectors to have width of less than 2.0 microseconds, and using a fast coincidence circuit to diminish pile-up artifacts. Preferably, the width is shaped to less than 0.5 microseconds and a very fast coincidence circuit is used to diminish the pile-up artifacts.

The signals from the detectors may be on-line or off-line analyzed by DSO to establish the fraction of pile-ups, and appropriate software correction is used to estimate the true count rate.

Methods can be implemented to eliminate temperature dependent drifts of count rate including the use of elements with low temperature coefficients, temperature stabilization and temperature compensation. The method may involve placing the scintillator in an appropriate Dewar and stabilizing its temperature electronically with a precision of a few degrees Celsius, preferably with precision of at least 1° C. This may involve use of both an appropriate heater (preferably ohmic heater) and cooler (preferably Peltier element) and

means to homogenize temperature by forced air flow. Temperatures of both scintillator and HVS/amplifier module may be sensed with precision of at least 0.2 ◦ C, and preferably with precision of about 0.1◦ C, by temperature to voltage converter (electronic thermometer) and the equivalent voltage is then measured by appropriate voltage sensor (ADC/DSO) in a central processing unit.

Preferably, an analog voltage signal equivalent to a temperature is sent to appropriate voltage sensor (ADC/DSO) in the central processing unit by means of shielded coaxial cable. The analog voltage signal equivalent to a temperature is preferably transformed in an optocoupler, sent by fiber optics, decoded by light to voltage converter and then measured by an appropriate voltage sensor (ADC/DSO) in the central processing unit. The analog voltage signal equivalent to a temperature is alternatively transformed in voltage to frequency converter, sent by shielded cable, preferably coaxial cable, and is measured by appropriate frequenciometer in the central processing unit. Preferably, measurement of temperature of scintillator and HVS/amplifier module is performed at least 10 times per second.

The method may involve implementation of a statistical rejection procedure. A preset number of temperature measurements (N) (typically a few hundred) is acquired, and temperature values that deviate from running average of temperature by more than 95% (more than two sigma) are rejected and replaced by weighted average over closest four measurements of temperature.

Preferably, the measurement of temperature of scintillator and HVS/amplifier module is used to calculate the true count rate, *i.e.*, the count rate drifts due to temperature changes are accounted for by software. Temperature compensation may be performed by means of calibration curve which is a product of three polynomial curves taking in to account temperature sensitivity of scintillator, PMT and HVS/divider and amplifier, respectively. Temperature compensation may use 3D look-out tables established in a precedent calibration procedure, or the temperature dependence of DSO is periodically measured with a system containing means for signal multiplexing and means to generate pulses of well known and temperature independent shape. Preferably, the temperature gain calibration for each detector is obtained by a series of measurements in both OR and AND mode acquisition.

A plurality of methods may be used to eliminate the long term drifts of count rate, *e.g.*, due to element aging, including the use of external sources of X-ray and gamma-ray photons with known energies with at least two different lines, including mechanical means for

removal or shielding the sources; and the use of two separate modes operation, namely measurement mode and calibration mode. At least three counters may be used in measurement mode and at least two channels of DSO in calibration mode.

A calibration mode comprises the steps of measuring pulse shapes, calculating the energy spectrum, estimating pile-up and dead-time corrections, and measuring external electromagnetic interference. The results of a calibration run can be compared with previous calibration runs, and the look-up calibration tables are upgraded for subsequent use for adjustment of raw data.

A plurality of methods can be implemented to compare the data in OR and AND data acquisition mode, including acquisition of at least 100,000 events with both pulse-height and pulse shape rejection enabled followed by the establishment of detection efficiencies in which the count rate rates in the detectors A and B are compared without the use of the above said rejections.

For fluids such as gases and liquids, and mixtures, steps may be taken to account for the influence of container walls, *e.g.* pipeline walls, including calibrations performed when the container is empty or filled with liquid of known density. Preferably, the liquids are either water or hydrocarbons with well known density.

Where the calibration source has activity lower than 0.5 microCi the energy spectrum is preferably obtained with DSO and all rejections enabled for three test configurations, *i.e.* with empty pipeline, pipeline filled with hydrocarbon with known density and pipeline filled with water. Where the calibration source has activity lower than 5 microCi the count rates in the selected energy window are preferably obtained with coincidence enabled for three test configurations, *i.e.* with empty pipeline, pipeline filled with hydrocarbon with known density and pipeline filled with water. Where the source has activity lower than 10 microCi the count rates in the selected energy window are preferably obtained with coincidence enabled.

A statistical rejection procedure may be implemented, wherein a preset number of coincidence counts (N) (typically a few hundred counts) is acquired, and the count rates that deviates more than two sqrt(N) from the average value N are rejected and replaced by weighted average over closest four measurements of count rate.

An apparatus according to the invention comprises

(a) a holder for placing a radioactive source near a sample so that photons from the source pass through the sample,

(b) at least two detectors capable of detecting at least two photons concurrently emitted by a radioisotope and generating a corresponding signal, at least one of the detectors being placed across the sample from the source and capable of measuring the attenuation and preferably the scatter of at least one of the photons concurrently emitted by the source, and

(c) a data processor for converting the signals to density measurements.

Using radioisotopes listed in Table 1, the detectors may be operated in coincidence mode in a symmetric sandwich configuration as in drawing 2; in a modified symmetric sandwich configuration as in drawing 3; in a compensated sandwich configuration as in drawing 4; or in a triangular configuration as in drawing 8; and for thin foil or plate samples, in a compensated flat symmetric sandwich configuration as in drawing 9, in an asymmetric sandwich with separator configuration as in drawing 10, or in a modified triangular configuration as in drawing 11.

Using radioisotopes listed in either Table 1 or Table 2, the detectors may be operated in coincidence mode in an asymmetric sandwich configuration as in drawing 5; in a shifted asymmetric sandwich configuration as in drawing 6; or in a modified asymmetric sandwich configuration as in drawing 7.

The source may be placed inside a spool made of material with appropriate mechanical characteristics (e.g. strength and weight) but low atomic number such as beryllium, plastic reinforced with appropriate fiber, aluminum, vanadium or titanium. Preferably, the spool has essentially elliptic cross-section, with total cross-section surface close to the cross-sectional surface of two pipes it is joining.

Alternatively, the source may be placed inside a bypass made of material with appropriate mechanical characteristics but low atomic number such as beryllium, plastic reinforced with appropriate fiber, aluminum, vanadium or titanium. Preferably, the bypass has essentially elliptical cross-section, with total cross-section surface much smaller than the cross-section of the main pipe.

The apparatus may further comprise a third, anticoincidence detector to diminish background as in drawing 12.

Preferably, the detectors are scintillators, and at least one of the detectors is NaI(Tl) with a thickness of at least 2" coupled to PMTs. In one embodiment, the detectors are of substantially the same size and consist of the NaI(Tl) scintillators with a thickness of at least 2" coupled to PMTs. At least one of the detectors may be a BGO scintillator with a thickness of at least 1" coupled to PMT. At least one of the detectors may be a BaF_2

scintillator with a thickness of at least 2" coupled to UV light sensitive PMTs. Or at least one of the detectors may be either GSO(Ce), CsF or CeF_3 scintillator with a thickness of at least 2" coupled to PMT. Where a radioisotope from Family 2 is used, at least one the said detectors is preferably a NaI(Tl), CaF_2(Eu) or YAP scintillator with thickness of less than 0.5" coupled to PMTs.

An apparatus optimized for radioisotopes from Family 2 uses hardware and software means to reject events due to interaction of CR in PMTs, and may have background due to electrons emitted from PMT diminished by application of appropriate thick optical windows made of quartz or other ultrapure, optically transparent materials. Alternatively, radioactive background is diminished by the use of an essentially cylindrical shield including at least one heavy metal component and placed around at least one of the detectors, and appropriate thick optical windows made of quartz or other ultrapure, optically transparent materials. The shield preferably comprises a few millimeter of Pb/Sn/Cu, the Cu being placed closest to the scintillator and further comprising a shield between the PMT and PMT base. One detector may be smaller than the other with the shield placed only around the smaller detector.

For radioisotopes from Family 2, the separator is preferably placed around the source and close to one of the detectors, and PMTs are selected to have radioactive background of less than 0.1 cps.

According to the invention, innovative use of MP-densitometry includes coincident detection and pulse shape analysis to achieve an excellent signal to background ratio (S/B) for low activity sources, and optimal use of photons with different energies. The following working definitions are used: low energy photons: 10 keV $<$ E $<$ 100 keV, and high energy photons: E $>$100 keV. Furthermore, photons emitted in the atomic shell rearrangements are called X-rays and photons emitted by the nuclear transitions and/or positron annihilations are called gamma-rays. The low energy photons (LEP) of importance in this invention are either X-rays or gamma-rays, but high energy photons (HEP) are always gamma-rays. To conclude this brief introduction we disclose a list of radioisotopes that are particularly useful in MP-densitometry, because they emit more than one photon. These fall into two main families:

Family 1: Positrons emitted by the source annihilate with a nearby electron into two gamma rays, each with an energy of 511 keV. These gamma rays are emitted back-to-back because of momentum conservation and their coincident detection is strongly angle-correlated. In positron-nuclear gamma (PG) emitters an emitted positron is followed by a

nuclear gamma transition. The positron annihilates, as described above and there is also an additional nuclear gamma ray. In nuclear cascade gamma (NC) emitters, a metastable nucleus cascades from a highly excited metastable state, and produces several gamma rays. The radioisotopes from the PG and NC groups that are important for MP-densitometry are listed in Table 1.

Family 2: Electron capture (EC) isotopes. In this case an atomic X-ray and a nuclear gamma ray are emitted (see Table 2).

Tables 1 and 2 show the lifetimes of the radioisotopes in years and days. The energies of the photons are listed in Handbook of Chemistry and Physics, Edition CRC, New York, relevant portion of which are hereby incorporated by reference.

Table 1. Isotope Family 1: Long life positron-gamma (PG) and nuclear cascade (NC) isotopes.

Na^{22} (2,58 y);
Al^{26} ($>10^5$ y);
Co^{56} (77.3 d); Co^{58} (71.0 d); Co^{60} (5.27 y);
Zn^{65} (245.0 d)
As^{74} (18.0 d);
Rb^{84} (33.0 d);
Y^{88} (108.0 d);
Tc^{95m} (60 d);
Rh^{99} (16 d); Rh^{102} (206 d);
I^{126} (13.2 d);
Eu^{152} (13 y);
Hg^{194} (130 d);
Bi^{206} (15.3 d)

Subtotal: 16 isotopes

Table 2. Isotope Family 2: Long Life EC isotopes.

Be^{7} (53.6 d);
Ar^{37} (34.3 d);
Ti^{44} (1,000 y);
V^{49} (330.0 d);
Cr^{51} (27.8 d);
Mn^{54} (291.0 d);
Fe^{55} (2.7 y);
Co^{56} (77.3 d); Co^{57} (270.0 d); Co^{58} (71.0 d);
Zn^{65} (245.0 d);
As^{73} (76.0 d); As^{74} (18.0 d);

Se^{75} (120.0 d);
Rb^{83} (83.0 d); Rb^{84} (33.0 d);
Sr^{82} (25.2 d); Sr^{85} (64.0 d);
Y^{88} (108.0 d);
Zr^{88} (85.0 d);
Tc^{95m} (60 d);
Rh^{99} (16 d); Rh^{101} (5 y); Rh^{102} (206 d);
Pd^{103} (17 d);
Ag^{105} (40 d); Ag^{108m} (>5 y);
Cd^{109} (470 d);
In^{113} (118 d);
Sn^{113} (118 d);
Sb^{119} (158 d);
Te^{118} (60 d); Te^{119} (45 d); Te^{121} (17 d);
I^{125} (60 d); I^{126} (13.2 d);
Ba^{131} (11.6 d);
Ce^{139} (140 d);
Pm^{143} (265 d); Pm^{144} (440 d); Pm^{145} (18 y); Pm^{146} (710 d);
Pm^{158m} (40.6 d);
Sm^{145} (340 d);
Eu^{148} (54 d); Eu^{149} (120 d); Eu^{150} (5 y); Eu^{152} (13 y);
Gd^{146} (48 d); Gd^{151} (120 d); Gd^{153} (200 d);
Tb^{160} (73 d);
Tm^{168} (85 d);
Yb^{169} (32 d);
Lu^{173} (1.3 y); Lu^{174m} (165 d);
Hf^{175} (70 d);
W^{181} (130 d)
Re^{183} (71 d); Re^{184} (50 d);
Os^{185} (94 d);
Ir^{189} (11 d); Ir^{190} (11 d); Ir^{192} (74 d);
Au^{195} (200 d);
Hg^{194} (130 d);
Tl^{202} (12 y); Tl^{204} (3.9 y);
Bi^{206} (15.3 y); Bi^{207} (30 y);
Pu^{237} (45.6 d);
Cm^{241} (35 d).

Subtotal : 72 isotopes

It is beneficial to be able to use a plurality of different MP-densitometer compatible sources. In some applications not only density but also composition need to be determined, *e.g.*, multiphase measurements in petroleum industry. This can be accomplished using numerous potential sources with emission energies properly selected to enable differentiation of the materials and to facilitate elimination of various backgrounds. Furthermore, sources with lifetime longer than a month are preferred.

The use of low activity sources is highly advantageous. The power and practicability of the disclosed procedures are connected with the use of new, very low background and highly sensitive detection techniques applicable to some radioisotopes. Specifically, the use of Multiple Photon Detectors (MPD) and appropriate radioisotopes from the families mentioned above are of crucial importance.

Very low radioactive background detectors (VLBD) are optimized for multiphoton radioisotopes, including positron-gamma (PG) emitters, nuclear cascade (NC) emitters, and electron capture (EC) emitters. Instruments for quantifying PG emitters were disclosed in United States Patent No. 5,083,026 entitled *Method, Apparatus and Applications of the Quantitation of Multiple Gamma-Photon Producing Isotopes with Increased Sensitivity*. United States Patent No. 5,532,122 entitled *Quantitation of Gamma and X-ray Emitting Isotopes* describes detection of EC radioisotopes. Such instruments are distinct from the use of multiphoton isotopes for densitometry.

In optimizing the performance of gamma densitometers the main challenge is to diminish the radioactive background and thus to achieve with a low activity source an excellent signal to background ratio, *i.e.*, S/B > 100, and high accuracy in density measurements. Basic elements that may be optimized are:

* the source;
* the detector; and
* the geometrical configuration of the system.

Other elements for optimization are:

* the coincidence trigger and on-line shape analysis;
* the temperature stabilization/compensation scheme;
* the calibration procedure; and
* the data acquisition and statistical procedures.

In the following we show that the innovative use of modified geometry, multiphoton coincidence and sophisticated signal analysis permits development of MP-densitometers with performances considerably better than prior-art gamma densitometers. System optimization depends on the sizes and densities of measured samples. These differences imply the use of photons with different energies, because the range of photons is strongly energy dependent.

In the case of PG emitters there are additional advantages due to the presence of back-to back annihilation photons, *i.e.*, the geometry is better defined for this case. Also, in PG emitters, additional third high energy photons permit redundant measurement, and allow

estimation of the average atomic number of the object under study. In the case of EC emitters there are typically only two coincident photons, and one of those is always a rather low energy X-ray (emitted due to atomic shell rearrangement). Thus, use of EC emitters leads to an asymmetric geometry, where two very different detectors are used. The NC emitters appropriate for MP-densitometry emit spatially non-correlated photons. When they emit two photons of very different energies, the instrumentation is similar to the case of EC emitters. The use of NC emitters is important because some of them produce two very high energy photons, like ^{60}Co with energies of 1,173 and 1,332 keV, respectively. In this case the geometrical configuration may be different from the case of PG emitters and both detectors can be located at the same side of the sample. Furthermore, one can use PG emitters where high energy photons, *e.g.*, annihilation photons are used in a coincidence scheme to reject background. Because very fast scintillators can be used to detect photons, important advantages of maximal count rate may be realized.

Each of these situations requires the use of photon detectors with different properties, leading to very different technical implementations. Thus for each of the cases the specific tradeoffs between the detection efficiency (DE), energy resolution (ER) and temporal response of the detectors should be selected. Furthermore, the photon energies considerably influence the choice and dimension of detectors. In the case of high energy photon (HEP) emitters, only heavy inorganic scintillators are practical and economical, whereas many different types of LEP detectors are available including scintillators, semiconducting and gas detectors.

Scintillators are the most popular detectors for quantitation of hard X-rays and gamma-rays. Good energy resolution is a major advantage in the design of low radioactive background detectors. Among the scintillators NaI(Tl) and CsI(Tl) produce the best light yield, while photomultiplier tubes (PMTs) permit counting of single photoelectrons. The NaI(Tl)/PMT combination is very popular, but other scintillator-PMT combinations are also of importance, including CsI(Tl), CsF, BF_2, CaF_2(Eu), YAP(Ce), BGO, and GSO(Ce). Here, we will differentiate between two classes of detectors appropriate for detection of soft X-rays (NaI(Tl), CaF_2(Eu)) and high energy photons (NaI(Tl), CsI(Tl), BGO, BaF_2, YAP(Ce), CeF_3, and GSO(Ce)), respectively.

Cost considerations should be emphasized. There are three elements which enhance practicability and diminish the cost of application of gamma densitometers:

* use of low level radioactive sources which do not require licensing;

* small size, portability and user friendliness; and
* low cost of the instrument.

Use of low level radioactive isotopes decreases the cost in three ways. First, the use of stronger sources requires licensed, specially trained operators and additional, often considerable, costs of transportation, reporting and waste disposal. Second, the stronger source requires larger shields and decreases the portability and user friendliness of the system. Finally, cost of isotopes increases and some of them are not available at larger amounts.

The MP-densitometer system should be portable, robust and user-friendly. Many of the operations are in remote locations, *i.e.*, system should be self-contained. Thus, low cost MP-densitometers which use scintillators were implemented. Furthermore, the read out electronics has been simplified and some functions which previously were implemented in hardware are currently accomplished in software. Particular combinations of hardware and software are disclosed.

The applications of MP-densitometry, especially in petroleum, chemical and nuclear industries require:

* good density resolution, say between 0.3 % and 3%;
* reasonably high dynamic range, say from 0.05 g/cc to 1.2 g/cc for gas/oil/water mixtures, and from 1 g/cc to 10 g/cc for solid samples;
* better than 0.5% reproducibility; and
* self-calibration and self-diagnostics.

The invention is further described below. In particular, means for achieving long term stability and repeatability of performance of MP-densitometers and advantages of different geometries and proper handling of temperature drifts and pileup artifacts are discussed.

DETAILED DESCRIPTION

In describing preferred embodiments of the present invention described here and illustrated in the drawings, specific terminology is employed for the sake of clarity. However, the invention is not intended to be limited to the specific terminology so selected, and it is to be understood that each specific element includes all technical equivalents which operate in a similar manner to accomplish a similar purpose.

The general principle underlying the present invention comprises a method according to which sources emitting concurrently a plurality of energetic photons (E > 10 keV) are

detected in setups with an optimized configuration. The apparatus implementing the optimal detection schemes are disclosed.

Standard gamma densitometer and MP-densitometer: Existing gamma densitometers use the simplest possible configuration: the sample is placed between the radiation source and the detector (see Figure 1). In this configuration only a few elements can be optimized; energy/activity of the source and the detector itself. In the following, we disclose a MP-densitometer that eliminates the limitations of prior-art gamma densitometers.

In many applications, current gamma densitometers are severely limited by the high level of required activity. For example, the Gamma-Trol densitometer produced by Tracerco Inc., England uses about 30 mCi source. The densitometers produced by other manufacturers have sources with similar activities. The thousand-fold background improvement achieved in MP-densitometers permits the use of a few microCurie sources, that are license exempt. Furthermore, the prior-art gamma densitometers often lead to artifacts in measurement. The external temperature change leads to count rate drifts which are misinterpreted as change of density of a measured sample. This problem is enhanced because gamma densitometers applied in petroleum production are often used in a desert environment, where diurnal temperature changes can be as large as 30-50°C.

Reproducibility and reliability are important features for gamma-densitometers. In many detectors, typical sources of uncertainty are self-absorption and errors due to variations in the source and detector positioning. Frequently used geometry is a flat scintillator coupled to a single PMT. Absorption artifacts are typically difficult to account for in this geometry. These artifacts can be considerably diminished when using two essentially identical flat detectors, each with independent readout electronics. We call such a configuration a "sandwich " detector, because both the source and the measured object are placed between the two detectors. When source self-absorption is negligible and the placement of the sample is correct, both detectors give essentially the same count rate. A sandwich configuration facilitates the use of highly efficient differentiation/compensation schemes.

The background due to Cosmic Rays (CR) is an important component of the total background in a single detector configuration. When two detectors are used, the energy deposited in one detector is often very different from the energy deposited in the second detector, which permits rejection of background due to CR primary particles. Also, as secondary particles shower initiated by CR in atmosphere have a characteristic density which considerably limits the sensitivity because of the background due to secondaries.

Furthermore, use of coincidence scheme is very efficient in eliminating radioactive background in scintillators, PMTs and shields.

Another important advantage of the sandwich geometry is its versatility. A preferred implementation of a detector according to the invention uses two essentially identical crystals. This is a good choice when annihilation photons are detected, and for some nuclear cascade isotopes, *e.g.*, ^{60}Co. However, many other important sources emit photons of quite different energies; often one photon is a soft X-ray ($E < 50$ keV) while the second photon is a nuclear gamma-ray, say $E > 500$ keV. This configuration is described in following section. In such case a sandwich detector consisting of two different scintillators with different crystal thicknesses is optimal.

Use of this geometry leads to double the number of elements, including PMTs which are a significant source of background. Furthermore, passive shielding is considerably more costly and the overall shape of the detector assembly is somewhat awkward and diminishes portability. Finally, the sandwich geometry and coincidence diminish the detection efficiency. Thus, the use of sandwich geometry is an innovative and rather counter-intuitive solution.

Conclusion 1: The prior-art gamma densitometers are limited by the use of single photon emitters. Use of multiphoton emitters permits the sandwich detector configuration and thousandfold lower background. This leads to improved signal/background ratio and permits high sensitivity with sources of low activity. Such MP-densitometers can be operated with a very low activity source, typically below 10 microCi, with better than 1% accuracy.

Diverse MP-densitometer configurations: MP-densitometers can operate in very different configurations as "symmetric" and "asymmetric" sandwiches, respectively. The use of sandwich geometry is counter-intuitive but enables the tradeoff between a lot of requirements because it:

* permits optimization of S/B ratio;
* permits operation in both non-coincident (OR) and coincident (AND) modes;
* permits to increase the sensitivity with respect to the sample density;
* minimizes attenuation artifacts; and
* diminishes many short term instabilities.

The simplest "symmetric sandwich" geometry is the one in which the MPD source is placed inside of the object whose properties are measured. This is the preferred implementation when properties of gas/fluid mixtures in flow are measured. The "symmetric

sandwich" geometry is shown in Figure 2. More specifically, for measurement of gas/fluid mixture density, a "spool" can be placed between two fragments of a normal pipeline. The "spool" has diameter equal to the main pipe but is made of material with low attenuation coefficient, *e.g.*, beryllium, plastic enforced with kevlar/carbon fiber, aluminum, vanadium or titanium. The "spool" is made according to stringent mechanical tolerances, *i.e.*, both the geometric dimensions and wall thickness are made with better than 0.1% precision. This facilitates the calibration of the MP-densitometer. This "spool" contains in the center a small, hydrodynamically shaped container in which the source is placed. This geometry is especially efficient and easy to calibrate in the case of PG emitters, wherein the two annihilation photons are emitted back-to-back. Thus, the count rates in both detectors should be very close to each other. This facilitates self-diagnostics and self-calibration.

The "symmetric sandwich" geometry was optimized for a particular situation of liquid flow. It allows measurement of laminar flow of gas/fluid mixtures, wherein the fluid moves close to the vertical pipeline walls and the flow of gas is mostly in the center of the vertical pipe. Note, that in this situation the velocity of gas and fluid are quite different leading to additional "dynamic pressure" compression of gas and modifying the efficient density. However, it can also be implemented in some other applications, such as in the measurement of properties of granulated material, *e.g.*, grain, seeds or cement.

For horizontal positioning of pipes, the "symmetric sandwich" geometry also permits determining if flow is turbulent (no difference between top and bottom detectors) or laminar (a few percent difference between top and bottom detectors). Furthermore, one can establish whether in the multiphase flow instabilities dominate, *i.e.*, if different phases of petroleum/water/gas mixture are separated. Another important feature of this configuration is that the total count is minimized and pileup artifacts diminished.

A somewhat more complicated "modified symmetric sandwich" geometry is shown in Figure 3. The advantage of this geometry is that it is applicable also in cases when the use of a "spool" is not practical. However, it retains the main advantages of a symmetric sandwich geometry, namely insensitivity to flow perturbation, improved pileup rejection and self-calibration capability. For example, because of overlapping absorption cones the effects of flow inhomogeneity can be diminished.

The "compensated sandwich" geometry is shown in Figure 4. Herein the source is placed between two objects; a measured sample whose properties are unknown, and a "standard sample" with similar and perfectly known properties. A pair of matched detectors is

preferred, with almost identical detection efficiency, energy resolution and temperature dependence. The concept of "relative" measurements is implemented, *i.e.*, difference in count rate is proportional to difference of densities between the "unknown" and "standard". This method is very efficient, when measurements of relatively small objects are required, especially when better than 0.1% precision in density/physical dimensions is needed. This configuration is "robust" and permits high reliability self-calibration. It is especially important, when the artifacts due to pileup and due to multiple scattering are important. Another innovative application is wherein the "relative density/dimensions" of an object are to be measured. By using the known standard, the majority of calibration artifacts, *e.g.*, temperature dependence can be eliminated.

We disclose special MP-densitometer configurations advantageous for large objects, say tubes with diameter larger than 6 inches. In such cases the "asymmetric sandwich" configuration shown is Figure 5 is the most useful. Herein, the PG source, *e.g.*, ^{22}Na, is placed close to a relatively small, say 2" in diameter fast scintillator. A very large detector, say 6" in diameter, is placed on the other side of the pipeline. Optionally, a big detector consisting of a plurality of smaller detectors operated in parallel may be used.

Note, that the large detector does not need to be placed exactly opposite the source at the other side of the object. This leads to a configuration called "shifted asymmetric sandwich" (see Figure 6). When the second detector is placed under an angle, higher absorption length is achieved, permitting better relation between density precision and statistical uncertainty in the count rate. The change of angle leads to some increase of absorption in pipe walls and increases considerably the absorption in petroleum. Thus, the efficient signal decreases and appropriate calibrations should be used. Furthermore, only part of the flow is sampled in density estimation. Actually, in this case absorption in petroleum is higher than absorption in the walls of pipe, and thedecrease in count rate should be comensated by the increase of measurement time. Once more, this configuration cannot be used in prior-art gamma densitometers which are essentially limited by their signal/background ratio. It is, however, useful in MP-densitometers wherein the sensitivity is limited by statistics rather than a signal/background ratio.

The "modified asymmetric sandwich" shown in Figure 7 is especially useful, when objects of very large diameter are studied using PG isotopes, including objects so large, that even annihilation photons are substantially absorbed. For example, for 10 inch pipelines with 0.375" steel walls about 95% of annihilation photons are absorbed. In this case, the two

detectors have very different functions. The detector close to the source detects unscattered photons and serves as trigger. It can be relatively small, say 2 inches in diameter. However, to diminish the pileup artifacts it should be made of a fast scintillator. The second detector measures the number and spectrum of scattered photons. It should be relatively large with size comparable to the thickness of the object to be studied. For example, in one of the implementations, the large detector consists of two slabs of relatively slow CsI scintillator, each about 2" thick and 16" long. Each CsI-scintillator is coupled to a separate PMT and chain of electronics. Using this configuration, the signal/background ratio better than 100 was achieved using only 1 microCi source for 10"-diameter pipes filled with water.

An important, "triangular" configuration is disclosed in Figure 8. This configuration is not applicable to PG isotopes but is appropriate for NC isotopes, wherein two high energy and directionally uncorrelated photons are emitted, *e.g.*, such is a case of ^{60}Co. This geometry is appropriate for density measurement in the largest pipes, wherein the highest possible energy of photons is an advantage because it limits the attenuation and permits reasonable statistics. Note, however, that only a fraction of the object volume is efficiently measured, which in the case of inhomogeneous objects including turbulent flow of liquids in pipes may lead to some artifacts. Data analysis minimizes these artifacts.

For large pipelines, a special "spool" can be produced from aluminum or titanium rather than stainless steel. For both materials, the stopping power is smaller for a thickness with the same mechanical properties. Thus, a few times lower attenuation in metal is achieved, and a lower activity source can be used. Furthermore, instead of a "spool" with essentially circular cross-section, the oval shape can be used. With appropriate dimensions, the change on the flow speed will be minimal but the attenuation will be a few-fold diminished.

Finally, the "spool" can consist of two parallel channels, *i.e.*, the liquid flow can be bypassed. Let us consider the 24" pipe with 1" walls. Assuming an 8" bypass the walls needs to be only 0.25" thick. Thus, the attenuation diminishes about fourfold. Taking in account the coincident method of readout used in the MP-densitometry; the activity of a source may be about 16-fold lower.

Conclusion 2: For different MP-densitometer configurations optimized for PG and NC radioisotopes, respectively, the performance advantages demonstrated in tests fully justify the use of these configurations.

MP-densitometer configurations optimized for EC sources: We disclosed above the MP-densitometers optimized for measuring densities of large (> 2" thickness) objects.

However, there are important applications for MP-densitometry for smaller objects, such as measurement of thickness of thin foils, flats and plates of plastic, paper and metal. In this case, the attenuation of gamma rays is small and sources with lower energy should be used. The MP-densitometers work best, when the sample absorbs from about 50 to 80% of emitted radiation. For the X-ray, however, the attenuation coefficient is very dependent on the atomic number of the absorbing media because the photoelectric effect dominates. Thus, the applicable energy range is from about 20 keV for paper/plastic/aluminum foils to about 100 keV for steel or cooper foils.

In our US Patent No. 5,532,122 and U.S.S.N. 08/669,970, the total counting rate is very small; typically less than 100 cps where S/B = 10, is typically judged adequate. Furthermore, in the biomedical applications of MPD, the attenuation by a sample is totally negligible and count rates in both detectors are practically identical. Also, the typical distance from one detector to another is very small and heavy shielding is often practical. In contrast, with MP-densitometry using EC sources:

1) the count rate is actually quite high, typically 100,000 cpm but excellent S/B > 1,000 is required;
2) the " asymmetric sandwich" or "triangular" configurations are advantageous and are used instead of "symmetric sandwich" configuration;
3) portability is an important issue, and the passive shielding should be considerably smaller than in stationary biomedical applications; and
4) matching of detectors properties is extremely important in MP-densitometry using EC sources.

Typically a measurement uncertainty of a few percent is required in prior MPD applications. It is mainly due to either radioactive background or statistical uncertainty; often only a few to a few hundreds of photons are collected. In MP-densitometers, *e.g.*, used to measure the thickness of thin layers, the measurement precision of 0.3% is typically required and 100,000's of photons are collected. The uncertainty is limited mainly by the systematic effects and artifacts. Thus, some further contrasts are:

a) in prior MPD systems the count rates in both detectors are identical, whereas in MP-densitometer the difference of count rate between two detectors is the signal;
b) influence of temperature drifts is negligible in MPD but is crucial in MP-densitometers;
c) the pileup and dead-time artifacts are very low in MPD but their elimination is over important in MP-densitometry; and

d) in MPD applications, the throughput is largely controlled by the user, *e.g.* measurement can almost always be repeated and time of measurement optimized, wherein in MP-densitometry any measurements interruptions (dead-time) are unacceptable.

Thus, even if some technical solutions enabling the low radioactive background incorporated into MPD and MP-densitometers are similar, the two devices are very different which forced a plurality of innovative modifications.

For small objects, excellent statistics can be obtained with a small activity source. Assuming that 50% of photons are absorbed in the object, and that the detection efficiency is about 50%, a 1 microCi source leads to a count rate of about 8,000 cps (50,000 cpm). This permits 1% statistical precision in density measurement for data acquisition time of a few seconds. However, at this count rate, pileup artifacts are not negligible and dead-time corrections important, making on-line pulse shape analysis difficult.

We designed and implemented the MP-densitometer based on use of low energy EC emitters. In the following, we call this device the X-ray densitometer. An appropriate EC source is ^{125}I, which emits photons with 27, 31 and 35 keV. For this source we achieved the radioactive background of less than 1 cpd. Thus background improvement of about 50,000 when compared with prior-art devices has been achieved. MP-densitometers optimized for the thin films application are of three different types, namely:

* "compensated flat symmetric sandwich" (see Figure 9);
* "asymmetric sandwich with separator" (see Figure 10); and
* "modified triangular" (see Figures 11 and 12).

Figure 9 provides a schematic view of a "compensated flat symmetric sandwich" configuration of a MP-densitometer. A single EC source is placed between two samples, one of unknown density/dimensions and another being a standard sample. Two matched detectors are used each with diameter much larger than the thickness of the objects. This is a geometry which facilitates the relative measurement and compensation of the artifacts providing the properties of two detectors are well matched. We expect that about 1.0% precision in thickness can be obtained for films with thickness of about 1 mm, and about 0.2 % for plates with thickness of about 1 cm. Herein, the EC isotope with very different energies of X-ray and gamma ray are used. The comparison of coincident rates on each side for different energies permits artifacts elimination. The use of a compensation scheme makes the measurement very reliable, *e.g.* if the foil from the same batch can be used as standard so that errors due to the variability in chemical composition of the sample can be eliminated.

A schematic view of an "asymmetric sandwich with separator" configuration of MP-densitometer is shown in Figure 10. A single EC source is placed on one side of the object whose density and/or dimensions are to be studied. Small X-ray detector is placed close to a source and larger gamma ray detector on the other side of the sample. The two detectors work in coincident mode. The advantage of this configuration is its versatility. Using it, objects of diverse shape, not only flats can be evaluated. Furthermore, the instrument can be relatively easily rescaled for samples with different dimensions. However, the attenuation properties of material from which the sample is fabricated has to be known rather well and a reliable calibration procedure is required. Actually, we disclose the use of an EC-densitometer in this configuration, where the dual or plural energy isotopes are used. By comparing the relative count rate at different energy, the attenuation properties of material can be calculated. However, the second detector should have the best energy resolution possible. Typically it will be a Ge detector or NaI(Tl) scintillator. Note the placement of the "separator" around the EC source. The use of separator permits substantial diminishment of cross-talk between the two detectors leading to low radioactive background. The use of a separator diminishes the requirements on speed of pulse-shape analysis and permits to overcome the pileup problem. This configuration may employ digital signal processor (DSP) cards.

The disclosed "asymmetric sandwich" configurations is enhanced by use of separators. In this case, one of the photons is an X-ray with relatively low energy and second photon is a gamma ray, whose energy can be selected to be quite high. Obviously, the soft X-ray cannot penetrate a large object but it can be used as a trigger to diminish the background. This configuration leads to the lowest background, because a X-ray detector can be very small and thin. Using this configuration, a background lower than 0.1 cpm was achieved. Furthermore, such detectors are much more portable because the X-ray detector can be miniaturized. From a large class of EC radioisotopes (see Table 2), the energy of a gamma ray can be selected to optimize the resolution of a MP-densitometer for a diverse object.

Figure 11 provides a schematic view of a "modified triangular" configuration of a MP-densitometer. A single EC source is placed on one side of the object whose density/dimensions are to be studied. Two gamma ray detectors are placed on the other side of the object. The size of detectors and the distance between them is much larger than the thickness of the object. The two detectors work in coincident mode and are separated by a passive shield. The main advantage of this geometry is that both detectors are placed on the

same side of a studied object. Actually, in many applications, the density of fast moving flats/plates is studied. The mechanical limitations of the transport system means that the volume under the flats is at premium. In this case the triangular geometry is advantageous because the EC source can be of very small dimensions, down to mm^3. Alternatively, the source can be a thin (< 1 mm in diameter), capillary or plastic rod. Another advantage of this geometry is that two detectors placed in parallel have much lower background that two detectors placed face-to-face. A factor of ten improvement in S/B is possible. Furthermore, in this geometry a sophisticated graded passive shield can be placed between the detectors. It can be further enhanced by the use of active anticoincidence detector as an anti-CR shield (see Figure 12). The disadvantage of triangular geometry is the lower geometric detection efficiency than other configurations.

When compared with previously disclosed configurations adequate for MP-gamma densitometers, a new element is the use of a "separator", a thin sheet of high atomic number metal with a hole in which the source is placed. The use of the separator, permits further background rejection. Note, that for high energy photons, the separator has to be so thick that it considerably limits the geometrical detection efficiency. For soft X-rays, the millimeter thick separator made of heavy metals (tungsten, gold, lead) absorbs almost all of the photons. It limits a crosstalk without significant change of geometrical detection efficiency.

In X-ray densitometer it is appropriate to physically isolate the scintillators as much as possible to reduce induced X-ray crosstalk between them. If the thickness of the samples is relatively small (a few millimeters) this can be achieved by incorporating a few mm thick sheet of lead or copper into the sample holder. Isolation of the detectors very effectively reduces the background in the single-photon ^{125}I region of interest (ROI). NaI(Tl) or CsI(Tl) based detectors show considerable sensitivity to the geometry and diameter of the opening in the separator. Detector isolation has less effect on the background in $CaF_2(Eu)$-based systems because these detectors do not produce secondary X-rays in the ^{125}I ROI.

Conclusion 3: Generally, the optimization of MP-densitometers for EC sources is a difficult, multiparameter process. It is highly sensitive to the properties of samples to be measured and the required measurement sensitivity. It is made even more complicated, because the sources of background are variable in respect to both time and location variable. Thus, the implementation of optimized MPD-densitometers using EC sources is not easy to model theoretically or numerically.

Uncertainty sources in gamma densitometry: The most important source of uncertainties is statistical limitations. In any measurement based on assembly of large number, N, of discrete measurements the statistical uncertainty is sqrt(N). Thus, in both gamma densitometer and MP-densitometer, at least 1,000,000 pulses are acquired to have the statistical uncertainty of about 0.1 % in a count rate. The uncertainty of density measurements is proportional to the uncertainty in count rate, but the coefficient is strongly variable with application. For example, in petroleum applications it depends on the pipeline diameter, thickness of the walls, distance between the detectors, and fluid density. In the case of MP-densitometry and using ^{22}Na source, the coefficient is 3, 2.5 and 2 for pipes with diameters of 3", 6" and 9", respectively. Thus, for 3" pipelines the statistical uncertainty in number of counts of 0.1% leads to density uncertainty of about 0.3%. Statistical uncertainty can be diminished by increasing either the source activity or the measurement time. In practice, the existing gamma densitometers are operated in a mode in which other sources of uncertainty are much larger than the statistical uncertainty. In contrast, the MP-densitometers have been optimized so that the statistical uncertainties have the same contribution to the overall error in density measurement as all other sources of uncertainties.

The main limitation of prior-art gamma densitometers can be traced back to:

* high radioactive background;
* short time instabilities, mainly due to pileup and dead-time artifacts; and
* temperature dependence of the count rate.

In prior-art detectors these sources of uncertainties lead to a few percent error in the density measurement. Note the complicated nature of the sources of uncertainty. For example, to overcome the high level radioactive background, the prior-art gamma densitometers use very strong sources (30 mCi or 1.1×10^9 decays per second). Even when assuming that only 1% of photons are detected, this leads to a 1.1×10^7 counts per second detection rate. At this rate, a pileup in a NaI(Tl) scintillator is above 50%, *i.e.*, the pileup deforms the shape and heights of every second photon. Thus, even rudimentary, pulse shape analysis is impossible. Even when using the faster BGO, about 10% of pulses pile up. In contrast, when using MP-densitometry, only 10 microCi source is used, leading to 1.1×10^6 decays per second. Taking in account the attenuation and the requirement of coincidence, counts rates are about 10^4 cps. At this level, pileup is still important in NaI(Tl) but totally negligible when using BGO, BaF_2 ,CsF or other fast detectors. Actually, even when using NaI(Tl), electronic means exist to minimize the influence of the pileup. The spurious signals

due to pulses overlap in the crystal itself can be partially accounted for by analyzing the pulse shapes for the selected sample of pulses; this method permits rejection of about 90% of pileup. Even with 10 mCi source in an appropriate configuration, the pileup and dead-time corrections are known to within 0.1 percent. However, this requires the availability of sophisticated on-line pulse shape analysis. In MP-densitometers we have been able to eliminate sources of uncertainty. The methods and technical details of pileup/dead-time corrections in MP-densitometer are disclosed in the following.

An important uncertainty in densitometry is a few percent overnight drift due to temperature sensitivity of system elements, mainly scintillators. In many applications the densitometers operate outside, *e.g.*, in the petroleum industry in desert conditions the diurnal temperature difference can be as large as 50 °C. In recent tests of the MP-densitometer in the CONOCO Test Facility at Lafayette, LA, a shadow of pipeline led to about 2 °C change in the temperature of the detector. Typically, the light yields of scintillators change about 3% for every 10 °C change of the temperature of scintillator, which in the above example would lead to about 0.6% uncertainty in density if measurements are performed with a standard gamma densitometer. Furthermore, the change of amplification in a PMT induced by temperature dependent drifts of high voltage and change of amplification in amplifiers have to be accounted for. In MP-densitometer, these uncertainties have been eliminated by methods disclosed in the following.

In petroleum applications, there are also sources of uncertainties due to absorption in pipe walls. Note that these limitations cannot be evaluated during manufacturing, but must be calibrated in field at the petroleum installation. For example, this is a case of limitations due to attenuation in steel of pipes. Herein, the mechanical tolerances on the dimensions and wall thickness are about 5%, wherein the density measurement with precision better than 1% is required. In case of MP-densitometry, the PG source, *e.g.*, ^{22}Na, emits not only two annihilation photons, but also 1275 keV photon. Comparison of the attenuation of these photons with and without fluid, permits to factor out the influence of pipe on the count rates. Furthermore, we observed that the mechanical precision of the placement of the pipe within the densitometer may lead to a systematic error of about 2-3% in the case of prior-art gamma densitometers. These kinds of errors are easy to detect and eliminate when using the MP-densitometer.

Conclusion 4: To overcome radioactive background problem, prior art gamma densitometers had used radioactive sources about 1000 times higher than used in MP-

densitometry. Thus, pileup and dead-time corrections are the main source of uncertainty in prior-art gamma densitometers, but can be minimized in MP-densitometers. When the background is pushed to below 10 cpm, the dominating sources of uncertainty are pileup corrections which limit the precision of NaI(Tl) based MP-densitometer. At that level, temperature corrections become dominating and innovative methods of their elimination used in MP-densitometers are disclosed.

Sources of background in Multi Photon Detector (MPD) systems: There are many sources of background and noise in nuclear radiation detectors. We believe that multiphoton based devices are the first very low radioactive background detectors (VLBDs) which can be reasonably priced (< $30,000) and small enough (10 x 10 x 50 cm) for general use in field applications.

We documented the following sources of background in scintillator-based gamma-densitometers and in MP-densitometers:

A1 radioactive contamination of detectors;
A2 radioactive contamination of PMTs;
A3 radioactive contamination of shields;
A4 high energy gammas from the environment;

B1 neutron induced gammas from the detectors, PMTs and shields;
B2 cosmic ray induced gammas from the detectors, PMTs and shields;
B3 direct hits due to cosmic rays;

C1 dark currents of PMTs;
C2 cosmic ray induced dark currents of PMTs;

D1 electronic pickup; and
D2 vibrational pickup.

Sources of background A1-A4 and B1-B3 are dependent on detector size; typically they grow linearly with the mass of the detector. Sources C1-C2 are dependent on the size of PMTs, and grow proportionally to their surface. The background sources D1-D2 tend to be independent of the size or type of the detector. Furthermore, they are much more important for photons with energy lower than 50 keV.

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The first group of backgrounds (A1-A4) leads to backgrounds of the order of a few cps. Also, the second group of backgrounds (B1-B3) is an important source of background in prior-art gamma densitometers. Actually, the background due to CR induced dark currents in PMTs (C2) are proportional to background due to a direct hit by CR (B3), which is much easier to count. Thus, in MP-densitometers, the background due to direct hits by CRs is periodically measured and used to evaluate the other sources of background (B1, B2 and C3). If the sum of these background is above a preset threshold (periods of high CR activity) they can be accounted for in the density measurement. The background sources from the third group (C1-C2) are the most difficult to reject by hardware means.

MP-densitometers use a synergistic combination of coincidence, hardware means and sophisticated pulse shape analysis. Coincidence permits about a 100-fold decrease in background, and additionally our pulse shape analysis software permits rejection of about 90% of the background. This requires use of on-line pulse shape analysis, including appropriate software. Software for background rejection limits the maximal count rate, and is very difficult to implement when the actual detector count-rate is much larger than 10 kHz. This limitation leads to highly innovative strategy of interleaving measurements and calibration runs.

The electronic and vibrational pickup noise is very much system and site dependent. In petroleum industry applications it tends to be large, because the detectors are placed close to pipeline, whereas the data acquisition system, *i.e.*, the computer, is often placed in a trailer. For example, during experimental tests of MP-densitometer in CONOCO Test Facility, Lafayette, Louisiana, this distance was about 90 feet. Even when using well-grounded coax cables, the observed electromagnetic noise without coincidence is about 100 cpm. In prior-art gamma densitometers, this source of noise is considerably lower than the radioactive background. However, in MP-densitometers wherein the radioactive background was diminished a thousandfold, the electromagnetic interference is an important source of uncertainty. We disclose the use of:

* coincidence counting wherein the signal from two detectors must arrive within 100 nsec;
* pulse shape analysis to evaluate the influence of electronic interferences;
* use of specially shielded BNC cables, or preferably fiber-optics, to diminish this source of background; and
* use of custom-designed low noise electronics.

All sources of background can be classified into two categories. First, there are sources which are essentially constant, *e.g.*, A1-A3 and C1. These sources of background can be accounted for by appropriate calibration, and their influence can be attenuated (but not eliminated). However, there are many sources of background which are highly variable, both geographically and as a function of time. For example, environmental radioactive background is often dominated by radon and is strongly weather dependent. Also, the background due to CR is not only geographic position dependent, but is also modulated by solar flares activity leading to changes of geomagnetic cutoffs. Thus, the radioactive background in prior-art gamma densitometers cannot be calibrated, leading to spurious residuals in density measurements. On the other hand, the background in MP-detectors is suppressed about a thousandfold and the background variations are negligible.

Conclusion 5: Very low background levels can be achieved at the earth's surface only through the synergistic use of a plurality of methods, of which coincidence and sophisticated pulse shape analysis are the most important. MP-densitometers are superior to prior-art gamma densitometers because all time/geographic location artifacts due to background are eliminated.

Implementations of MP-densitometers: In MP-densitometers according to the invention, the coincidence between two photons emitted by an appropriate source is used to diminish the radioactive background. For example, when using a suitable long life PG source (^{22}Na), scattering of the annihilation photons leads to change of both total number of detected photons and to substantial change of the spectrum. Algorithms for estimating the density of the studied object depend on this change of the spectrum. Thus, the use of scintillators with the highest energy resolution, namely NaI(Tl) is disclosed. However, for larger objects, this leads to either reduction of detection efficiency when 3" thick crystals are used or increase of background when 4" or 5" -diameter crystals are used. For example, in densitometry of petroleum products in flow, the optimal NaI(Tl) crystal diameter is a function of pipeline diameter. For pipelines with a diameter up to 6", the 3" crystals are optimal. For 10" and 16" pipeline diameters, the optimal crystal diameter is 4" and 5", respectively.

The rise-time of scintillation light in NaI(Tl) is about 200 ns, and typically about 500 ns pulse shaping is used. For sources in excess of 5 microCi this leads to the pileup artifacts. We observed the uncertainty of about 0.5% in count rate for 3" pipelines filled with petroleum. This leads to 1.5% precision in density measurement. However, the use of the

geometry shown in Figure 3 with each source about 5 microCi diminishes by a factor of two pileup and permits a better than 1% precision in density measurement. Furthermore, the uncertainty in count rate has a diminishing influence on precision of density measurements for larger pipes, *e.g.*, for 6" pipe the density measurements uncertainty of about 0.5% is possible with two 5 microCi sources.

MP-densitometers may be improved by the use of scintillators different from NaI(Tl). For example, CsI(Tl) has only marginally lower energy resolution but about twice higher stopping power. Alas, it is a slower scintillator than NaI(Tl) and thus leads to higher pileup artifacts; CsI(Tl) should not be used when the source activity is above 1 microCi. Use of BGO is suggested, even if this crystal has about four times lower energy resolution. The advantages of BGO crystals are about three times higher stopping power and faster scintillation. Thus, for the same stopping power the pileup is about a factor ten smaller and also background is somewhat lower. Finally, when a very high precision of density measurement is required, say 0.1%, the higher activity sources may be used to provide the appropriate statistics. In this case even in the symmetric sandwich geometry, the errors are mainly due to pileup. To diminish pileup, the fastest detector with appropriate stopping power should be used. Thus, we disclose the use of BaF_2 GSO(Ce), YAP(Ce), CsF, and CeF_3, which are ten times faster than BGO and a hundred times faster than NaI(Tl). Note, that if the pileup is not critical, the use of BaF_2, GSO(Ce), YAP(Ce), and CsF is highly counter-intuitive because it features lower stopping power, worse energy resolution and requires very costly quartz PMTs.

In MP-densitometers using EC sources, the X-ray detector may be built of CaF_2(Eu) rather than NaI(Tl). Using CaF_2(Eu) some sources of background (especially the dark currents) can be rejected better than when using NaI(Tl). Alas, CaF_2(Eu) is also a much slower scintillator, and typically the shaping time of a few microseconds is used. Thus, it should be used only when the radioactive source is below 0.5 microCi, *i.e.*, only in the case of rather small objects.

Finally, we disclose the use of yttrium-aluminum-phosphate scintillator (YAP(Ce)) instead of NaI(Tl) as a X-ray detector in an asymmetric configuration of the MP-densitometer. YAP(Ce) is not-hygroscopic and is more robust mechanically than NaI(Tl). Second, it is a very fast scintillator and pileup is negligible even for 10 microCi sources.

Conclusion 6: We disclose the use of a plurality of heavy inorganic scintillators in construction of MP-densitometers. Their advantages and limitations in diverse applications

are discussed. More specifically, the utility of fast scintillators (YAP(Ce), BGO, BaF_2, CsF, and CeF_3) is disclosed.

Temperature stabilization/compensation schemes in MP-densitometers: Accurate measurement of density requires a high stability detector system. The observed drifts are predominantly due to changes in ambient temperature, which cause drifts in the yield of the scintillators as well as changes in gains of the PMT's and amplifiers. In prior-art gamma densitometers, the temperature drifts are the dominating sources of measurement errors. For example, many oil wells and pipelines are located in deserts, where the diurnal temperature variations can be as large as 50°C. Similarly, the temperature gradients are very large and site dependent in the metallurgic industry where, the gamma densitometers are often placed very close to moving slabs of very hot metal.

Typically, the light yield from NaI(Tl) scintillator is changed by up to 3% when temperature varies by 10°C. The typical dependence of a coincident count rate in NaI(Tl) based MP-densitometer on temperature is shown in Figure 13. For majority of scintillators including NaI(Tl), the light yield is a monotonic function of temperature and decreases for lower temperature. However, the temperature dependence of scintillator may be more complicated in particular cases. For example, in BaF_2 there are two components, one very fast emitting an UV light and another much slower emitting in visible. The first component is almost temperature independent, wherein the second component is strongly temperature dependent and becomes negligible at above 65°C. Preferably, with BaF_2 in MPD-densitometry, the crystal temperature is kept above 40 °C to diminish artifacts.

The main method of correcting for temperature artifacts is to stabilize the temperature of the crystals. Each detector may be independently stabilized in temperature. The schematic drawings of the temperature stabilization system are provided in Figure 14. For example, in the case of petroleum industry, each detector is placed in a massive, explosion proof box typically made of aluminum. The appropriate Dewar with internal dimensions of about 3.5" is mounted inside this box. PMTs are very bad heat conductors due to a high vacuum inside and only a very thin wall of glass. Thus the Dewar covers essentially only a scintillator and PMT assembly and is additionally insulated around the PMT neck with porous plastic material. The fast reacting ohmic heaters, *e.g.*, high power transistors are placed on the good conductor metal plates, typically aluminum or copper attached to the crystal. The temperature is homogenized inside the Dewar by the use of miniaturized fans. The temperature stabilization is obtained by the direct feed back using a fast electronically

readable thermometer. We also disclose a somewhat more sophisticated temperature stabilization system in which all elements of stabilization system (heaters, fans, thermostats) are computer modifiable. A feedback loop is established between the output of scintillators (position of photoabsorption peak) and the parameters of the temperature stabilization system. Practically, it is preferable to use only heating rather than both heating and cooling. In this case, the temperature is established higher than the expected maximum temperature of the environment.

Such a temperature stabilized system permits operation at an essentially stable temperature regime, where the temperature drifts and fluctuations are lower than 1°C. However, even this temperature stabilization is not sufficient for 0.1% count rate stability. Thus, the additional temperature compensation system is used in MP-densitometer (see Figure 15). Herein, three temperature sensors are placed in each detector; two on the crystal and one on the integral high voltage supply (HVS)/amplifier module. The thermometers output is amplified by a DC-DC amplifier to a signal of about 5V. The information from these sensors is sent several times a second to a digital oscilloscope card and is on-line analyzed by the computer. We disclose the use of multiplexing system, wherein the outputs of the three thermometers are sequentially inputted on DSO. To diminish the electronic interference, the analog signal is sent by well-shielded BNC cables.

Optionally, the voltages from electronic thermometers are electronically modified in a voltage-to-frequency converter, and are sent by the cables that are used for transmission of pulses from detectors to data acquisition computer. In yet other implementation, the electronic thermometer has a digital RS320 interface and is directly coupled to the input ports of the data acquisition computer. After reception the data are software filtered to remove spikes due to electromagnetic interference and averaged. In all these implementations the reliable temperature measurements with precision of about 0.2% were achieved. Note, that the temperature of crystals is often a few degree Celsius different from temperature of PMT base. This is accounted in software in which three temperature compensation curves are calculated independently, for scintillation crystals, high voltage supply/dividers and signal amplifiers, respectively.

Both heating and cooling by means of the Peltier effect may be used so that the crystals temperature is stabilized below the ambient temperature. This permits use of the operational temperature, in which the temperature coefficient of the scintillator is minimal.

The temperature dependent drifts of the high voltage supplies, preamplifiers and ADC's are important, and it is preferable to use stable electronics and high voltage power supply for the PMTs. The high voltage power supplies for the PMTs have been stabilized by introducing a high gain negative feedback and using as a reference Max 580 chips with low temperature coefficient (1-2 ppm/°C). We also use 1% metal film resistors with a low temperature coefficient. The voltage divider for the PMT is based on the same type of resistors. The temperature dependence of the preamplifiers has been minimized by using highly stable operational amplifiers and distributing the gain so that each amplifier cascade has a gain lower than five. The result of the work of temperature compensation scheme is shown in Figure 16, where the row data, and corrected data of overnight measurement are plotted.

The temperature sensitivity and long term stability of DSO's are especially important in coincident systems. The relative drifts of two channels as a function of ambient temperature change is often larger than the intrinsic resolution of DSO. Thus, we disclose innovative controls for our MP-densitometers. First, at the known repetition frequency (typically every few minutes), the two detectors are commuted into two channels of DSO. This permits us to distinguish if long term drifts are due to scintillators/front electronics or data acquisition, especially ADC's. Second, after each measurement period, the calibration procedure is initiated, wherein the pulse from the well-stabilized pulse generator is applied in parallel to both channels of a data acquisition system and counted in both OR and AND modes. Actually, to check for possible nonlinearities of data acquisition, the pulses of diverse amplitude are counted and their shapes registered.

Conclusion 7: Advanced temperature stabilization/compensation systems can be used in MP-densitometry. The temperatures of the crystals are stabilized within a 1°C by active temperature control system. Additional and independent electronic systems measure the actual temperature of the scintillators and HVS/amplifiers with precision better than 0.2 °C. Software permitting on-line compensation of the count rate is disclosed.

Two modes of operation of MP-densitometers: The applications of MP-densitometers can be divided into two classes. In the first class, density measurement with precision of a few percent is required and long term stability of the count rate should be about 0.3 to 0.5%. The above described techniques of temperature stabilization and compensation are fully sufficient. Furthermore, a periodical calibration of detectors is recommended.

In the second class of applications, *e.g.*, in two phase measurements of oil/water mixtures, better than 1% density precision is required. Thus, the count rate uncertainty should be 0.1 - 0.3% which is really difficult to achieve. To enable this count-stability a few innovative techniques are used in MP-densitometers to further diminish the remaining temperature induced drifts in the PMTs and scintillators. These drifts can be compensated by adjusting the gains of the amplifiers, so that the acquired spectra will be unchanged. Electronics permits control of these gains. When coupled with temperature readout devices placed in the detector system this allows adjusting the gains to compensate for temperature induced drifts. The temperature-to-gain calibration for each detector can be efficiently obtained by a series of measurements in non-coincident (OR) mode and use both OR and AND mode acquisition. The entire calibration procedure is automated and can be periodically performed to compensate for long-term drifts.

An optional element of the long term stabilization of the detectors is the use of calibrated low energy photon sources consisting of at least two low energy lines. These calibration energies are sufficiently different so that these two lines can be distinguished by a detector. Preferably, this is a few hundred nCi source, that is mechanically shielded and the obscurator is computer activated.

Therefore, the MP-densitometers use two modes of data acquisition: measurement mode and calibration mode. In measurement mode the main effort is to diminish the pileup and dead-time effects. Thus, the fast electronics and system of fast counters are used. In the calibration mode, the main effort is to evaluate the influence of different background sources (natural radioactivity, CRs, electronic interference). Also, the efficiency of temperature stabilization/compensation system is checked. In calibration mode, the pulses from the counters go to a fast digital storage oscilloscope (DSO) working in the sampling mode. These data are dumped into computer memory and processed in parallel with data acquisition. The software performs the following steps:

* measuring the pulse shapes;
* calculating an energy spectrum;
* estimating the pileup and dead-time corrections; and
* measuring the external electromagnetic interferences.

From such short calibration runs, the look-up calibration tables are upgraded which are used to adjust the raw data. In calibration mode the data are obtained for both detectors in

OR as well as in AND modes. Thus any differences between the detectors showing a malfunction can be analyzed.

The MPD normally operates in OR and the AND mode, with considerable background reduction through pulse shape analysis. In a 3" diameter and 3" thick NaI(Tl) crystal, the typical background without shape analysis is about 100 counts per second. Using the pulse shape analysis, in MPD the background in the OR mode is 50 counts per minute. In AND mode, the MPD instruments permit about thousandfold lower background. During the data acquisition both non-coincident and coincident events are identified and counted. Upon completion of the acquisition the MPD outputs both OR and AND counting data. The AND data serve to establish the material density and the OR data are also analyzed (either on-line or periodically) to permit the reliable calibration of the system, *e.g.*, to monitor the temperature drifts and pile up problems.

The software estimates the pileup corrections to the counting rate. These corrections are very different at OR and AND mode of operation. It is important that the pileup corrections are estimated at the level of raw data, *i.e.*, before background rejection. Self-diagnostics and self-calibration programs permit concordance of the OR and AND counting rate data with accuracy of about 0.2%.

In an automatic device calibration/ROI setting algorithm, the calibration procedure depends on the MP-densitometer configuration, and in the following the case of "modified symmetric sandwich" using ^{22}Na source, is described as an example. The user can request the software to perform this procedure at any time but typically the calibration runs are performed each hour. No calibrated source is needed, because the reasonably high activity (1- 5 microCi) ^{22}Na source is already present. The program acquires 100,000 events with all pulse-height and pulse-shape rejections enabled. Once the acquisition is finished, the spectrum is analyzed to determine the ROI for a photoelectric absorbtion peak of annihilation photons. The count rate (cpm) within this ROI is determined. As the actual activity (dpm) in the source is known, the detection efficiency may be established from the ratio of cpm/dpm. To estimate the absolute activity of the calibration source, the program starts a second round of data acquisition with pulse-shape rejection disabled (as the calibration source activity is high at the 511 keV line, there is no need to reject the background, and no real events are discarded). Spectra for both detectors (A and B) are built until 200,000 events are acquired. Subsequently the spectrum of each detector is analyzed to estimate the count rates in the 1-photon and 2-photon peaks and efficiencies of the detectors are calculated. The

data are checked for consistency by comparing the estimates from detectors A and detector B. Also, the pileup pulses are detected and their frequency estimated. With the pulse shape analysis available, the pileup pulses are easily detected due to a characteristic, two peak structure. With statistics of about 200,000 pulses analyzed the sub-set of about 2,000 pileup pulses are typically analyzed, for which the distance between two peaks is measured. This permits statistical analysis of the pileup artifacts.

After the autocalibration/ROI setting is complete, the system is ready to count samples of the same geometry as the last calibration sample used. Acquisition is performed either for a preset time or until a preset number of counts (determining the statistical uncertainty of counting) have been acquired in the ROI. The program then estimates the actual decay rate in the source using the previously computed DE. The sample counting data are then stored to disk in an ASCII data file which can be transferred to any database or spreadsheet program for analysis.

If possible, the calibration is performed with pipelines empty or filled with water so as to permit comparison with previous calibration runs. However, the calibration can be performed with any fluid in the pipeline because it is mainly based on pulse shape analysis of non-attenuated annihilation photons in OR mode.

Conclusion 8: In a calibration mode using pulse shape analysis based on a digital storage oscilloscope, interleaving the measurement and calibration runs permits improved background understanding. It also permits better control of artifacts due to pileup and temperature induced drifts. The calibration methods are highly computer intensive.

EXAMPLES

Specific embodiments of the invention will now be described in more detail, by way of example only.

Comparative tests of MP- and standard densitometers: An MP - densitometer for measurement of gas/oil/water mixture density in the density range 0 - 1.05 g/cc was tested both in the laboratory and in a CONOCO Test Facility at Lafayette, LA.

The two detectors with 3" NaI (Tl) crystals were coupled to 3" PMT and bases (HVPS/amplifier/shaper) and assembled inside explosion proof aluminum boxes. The signals were processed by amplifier/shaper electronics and then transferred to a PC 486/66 equipped with proprietary data acquisition cards. The computer was installed in the control room about 90 feet away from the detectors. The coincident trigger selected the coincident signal

from two detectors within the energy interval 100 - 600 keV, which correspond to both scattered and not scattered annihilation photons.

The MP-densitometer is working in three modes: in two calibration modes and in the data acquisition mode. In the first calibration mode the signal from a low activity (0.3 microCi ^{22}Na) source is processed by a proprietary coincident card and is subsequently analyzed with the DSO. The coincident trigger is tuned for a selected region of interest (ROI) in energy. The ROI depends on a geometrical configuration of the system and may be different for two detectors in an asymmetric configuration.

For density measurements the MP-densitometer is calibrated using the 2 microCi source in two density points: with an empty and water filled pipe. In this mode the smoothed count rate and temperatures (see below) are stored and temperature compensation look-up tables are calculated.

In data acquisition mode (with 2 microCi source) the signal from a coincident trigger is accumulated for a given number of times (100 times per minute). The use of a statistical rejection procedure is disclosed, wherein the count rates that are more than two sqrt(N) off from the average value N are rejected and replaced by a weighted average over the closest four points. Then all the counts are summed over and this number is accepted as a real number of counts during a given time interval (1 min). The same procedure is applied to the signals from two temperature sensors installed on the two detectors. Every minute the signals from the temperature sensors, coincident (AND) count-rate and non-coincident (OR) count rates are written in the file. The density of the mixture inside the pipeline is calculated and stored using the coincident count rate corrected for the current detector temperature, and the reference points obtained during the calibration run. The time dependence of the temperatures of both detectors, coincident count rate and density are presented on the monitor in the real time. The digital output density signal is converted into analog DC voltage in the range 1 - 5 V.

Selected results of the tests are presented in the Figures 17-19. In Figure 17 the results of measurements of oil/water mixture density for different fractions of oil in the salt water are shown. The density was measured by MP-densitometer installed at the vertical 3" pipe two feet below Gamma Trol densitometer using 28 mCi source of ^{137}Cs. The density was averaged every 60 seconds by both densitometers. The density of oil and water were measured separately in before mixing by Solartron densitometer with precision better than 0.1%. The densities of oil/water mixtures calculated on the basis of these measurements and

the readings from CONOCO Test Facility flowmeters are shown in Figure 17 as the data by Solartron. The precision achieved for 60 second measurement time by the prototype of MP-densitometer was comparable to the one achieved by Gamma Trol densitometer, which used 15,000 more active source. On the other hand the systematical uncertainties of Gamma Trol densitometer in the region of low water fractions are substantially higher than these of MP-densitometer. The same data after averaging during 4 minute intervals are shown in Figure 18. In this case the uncertainties of MP-densitometer with 2 microCurie source are close the uncertainties of Gamma Trol densitometer.

The results of measurement of gas/water mixture density are shown in Figure 19. In this case the precision of measurement is not determined by statistical uncertainties, but by systematical uncertainties of a few percent due to turbulent flow of gas/fluid mixture in the pipe. Both densitometers gave close results for density measurements and for both densitometers even for one minute measurement time the precision of density measurements was determined by the systematical uncertainty.

Conclusion 9: The use of MP-densitometers permits comparable or better density measurements than prior-art gamma densitometers even if 15,000 lower activity of source is used.

MP-densitometer optimized for EC sources: There are important applications of MP-densitometry for smaller objects (< 3"). Such as the measurement of thickness of thin foils and slabs of metal, plastic and paper. In this case, the attenuation of gamma rays is small and sources with lower energy should be used. The MP-densitometers works best of all, when the sample absorbs from about 50 to 80% of emitted radiation. For the X-ray, however, the attenuation coefficient is very dependent on the atomic number of the absorbing media because the photoelectric effect dominates. Thus, the practicable energy range is from about 20 keV for paper/plastic/aluminum foils to about 100 keV for steel or cooper foils.

The appropriate configurations are the "compensated flat symmetric sandwich " (see Figure 9), "asymmetric sandwich with separator" (see Figure 10), and "modified triangular" (see Figure 11). The relative advantages of this configurations have been discussed above. In the three detector configuration (see Figure 12), the third, anticoincidence detector is used to eliminate the cross-talk between the two signal detectors.

For small objects excellent statistics can be obtained with a small activity source. Assuming, that 50% of photons are absorbed in the object, and that the detection efficiency is about 50%, the 1 microCi source leads to a count rate of about 8,000 cps (50,000 cpm).

This permits the 1% statistical precision in density measurement for a data acquisition time of a few seconds. Alas, at this count rate pileup artifacts are important. Thus, the software based pulse shape analysis using DSOs and subsequent software analysis is not adequate and the background due to CR induced cross-talk between detectors will dominate. To implement the cross-talk rejection in "compensated flat symmetric sandwich " and "asymmetric sandwich with separator" we disclose the use of a fast digital storage processor (DSP) based pulse shape analysis.

Generally, NaI(Tl) scintillator crystals are believed to be optimal for detection of X-rays. In coincident mode, the background rejection is inversely proportional to the square of the energy resolution, which for NaI(Tl) is about 50% better than for other scintillators. Furthermore, among scintillators with reasonable energy resolution (NaI(Tl), CsI(Tl), YAP(Ce) and CaF_2(Eu)), the YAP(Ce) and NaI(Tl) scintillators are the fastest. Once more, according to prior-art practice, background rejection is proportional to the square of the timing resolution. The theoretical estimates suggest that the background in an MPD based on NaI(Tl) scintillator should be about four times lower than when using other scintillators. In practice, an MPD system can achieve a background of about 0.5 counts per week (0.5 cpw).

However, we also observed some real inconveniences of NaI(Tl) based devices:

* NaI(Tl) is mechanically fragile, *e.g.*, it often cracks when submitted to temperature gradients and/or during transportation;
* NaI(Tl) must be hermetically sealed, and the thin Al or Be foils covering the front surface of the scintillator is easy to rip off when samples are placed in its vicinity; and
* when samples of larger diameter are used, the background in MPD systems based on NaI(Tl) deteriorates rather fast due to crosstalk between the crystals.

For example, there are some changes in NaI(Tl) crystal properties over a 2-year period in about 40 NaI(Tl) crystals which we have tested. Some (about 10%) crack from thermal stresses while others (about 15%) are turning yellow due to their hygroscopicity. In contrast, no significant variability in crystal properties was observed for twenty CaF_2(Eu) crystals over one year interval.

A series of measurements in which for soft-X rays NaI(Tl) was replaced with CaF_2(Eu) scintillator confirmed our theoretical prediction that a simple replacement leads to a factor of few increases in background. However, following a step-by-step process of innovative changes in readout electronics and after developing an additional 10,000 lines of software,

we have achieved considerable improvements. Currently, $CaF_2(Eu)$ and NaI(Tl) based MPD systems have comparable background.

$CaF_2(Eu)$ has a surprisingly low background for ^{125}I detection because in AND mode the background is dominated by soft X-ray emitted and absorbed in one crystal coincident with some source of energy detected in the second crystal. When NaI(Tl) is used, any absorbtion of an external photon with $E > 35$ keV in the crystal leads to remission of 26 keV or 32 keV photons from rearrangement of atomic shells. Thus, the characteristic iodine X-rays are emitted when the NaI(Tl) crystal is used. These cannot be distinguished from the 25 and 31 keV photons emitted by ^{125}I. Fortunately, $CaF_2(Eu)$ consists of only low atomic number elements. Thus, the characteristic X-rays have less than 15 keV energy and can be differentiated from radio-iodine X-rays. Thus, as a rule of thumb, NaI(Tl) or CsI should be used for EC radioisotopes with atomic number of either less than 40 or larger than 70. For EC isotopes with atomic number between 40 and 70, $CaF_2(Eu)$ is a preferred scintillator.

In MP-densitometer using EC sources the crystals themselves are preferably very thin and the surface of the PMT cathodes is about fifteen times larger than the surface of the crystals. A high energy CR striking one of the PMT anodes gives rise to an avalanche of electrons which are subsequently amplified. Such pulses lead to an apparent energy deposition much lower than the energy of the CR, *i.e.*, there is a considerable overlap between the energy spectrum of CR induced dark current pulses in the PMTs and the energy from radio-iodine. In MP-densitometers optimized for EC sources and using scintillator-PMT combinations this source of background is quite important. Fortunately, the energy deposited in the scintillator typically leads to pulses longer than the CR induced pulses in the PMT, which has a characteristic time constant of less than a nanosecond. The difference in pulse rise times between CR induced PMT pulses and those created in $CaF_2(Eu)$ is large; 0.1 ns and a few microseconds respectively. In an embodiment of a $CaF_2(Eu)$ based MPD system, about 95% of CR induced PMT artifacts are rejected on-line.

For low-background counting the scintillators used in X-ray detectors should be shielded from radioactivity in the photomultipliers to which they are coupled, such as from beta particles and low energy photons. For example, a 5-mm thick quartz window is placed between the PMT and the scintillator. Quartz is preferred both for its excellent optical properties and its high purity; quartz matches the optical density of $CaF_2(Eu)$ very well and is acceptable for NaI(Tl). We did not observe any increase in background due to radioactive contamination in quartz. Further improvement can be achieved by using materials with

higher stopping power than quartz. We disclose the use of high-purity GeO_2 and germanium-based glasses for this purpose. They have very low intrinsic radioactive background and their higher atomic number and density is an advantage over quartz windows. Such windows with a few millimeters thickness will efficiently stop low energy photons as well as beta particles without degrading the optical qualities of the scintillator/PMT systems. Actually, the optical properties of gelica and germanium glass match NaI(Tl) better than quartz does. Even higher density glasses based on lead may be used as well as high density transparent crystals such as PbF_2 and bismuth germanite (BGO). In the case of BGO, undoped crystals should be used so as not to generate artifacts due to scintillation within the BGO. The optical density of these materials is higher than for CaF_2(Eu) or NaI(Tl). The use of a thin layer of special optical greases, *e.g.*, powdered PbF_2 in silicon grease can match the optical properties of the scintillator and window and the window and PMT.

Both background and detection efficiency in the modified asymmetric sandwich geometry depends on the dimensions of the small crystal. For many applications the source can be small, often about 1 mm in diameter. An optimal configuration is one with two crystals of both different material and different diameters. The detector close to the source should be small and thin, typically 0.75" or 1" in diameter. However, the detector on the other side of the measured object should be much larger, typically 3" or 4" in diameter. In a preferred design of MP-densitometer using EC isotopes, the two detectors are differentiated not only by their dimensions but also use different scintillators; depending of the type of source used it should be either NaI(Tl), YAP(Ce), CsF or BGO.

A preferred implementation of the MP-densitometer optimized for detection of EC isotopes, *e.g.*, ^{125}I, is an "asymmetric sandwich", consisting of two modules, the smaller of them preferably placed in a low radioactive background shield consisting of heavy metals, typically a sandwich consisting of a few millimeters of Pb /Sn/Cu. The shield is essentially cylindrical, with the thin window close to where the scintillator is placed. Also, there is a thinner shield between the PMT and the PMT base containing high voltage supply, high voltage divider, and preamplifier. Additionally, the somewhat thicker shield is placed after the said PMT base.

Each of the detector modules consists of the following elements:

* inorganic scintillator with thickness optimized for a given emitter;
* high purity optical window, placed between scintillator and PMT;
* selected, low radioactive background PMT;

* a graded passive shield between the PMT and PMT base assembly; and
* PMT base assembly consisting of HV supply, high voltage divider and preamplifier.

^{125}I is an appropriate source for a plurality of MP-densitometry applications, preferably using flat detectors (1 mm thick NaI(Tl) or 1.5 mm thick $CaF_2(Eu)$). Thinner scintillators decrease the DE of the system while for larger crystals the signal to background ratio effectively diminishes. The scintillators are coupled through quartz windows 3-5 mm thick to a high resolution PMTs which are selected for low background. The PMT signals are amplified and shaped using proprietary electronics built into the PMT bases. To reduce the flux of background photons from the bases to the scintillators the bases are isolated from the PMTs with 5 mm of lead and 1 mm of copper plating with holes for the PMT pins.

At least one of the detectors is placed in graded lead+tin+copper shields (typically, 0.25" lead, 2 mm tin, 1 mm copper). The detectors are placed face-to-face a few millimeters from one another and a crosstalk eliminator is placed between one of the detectors and the sample. Typically, this is a 1 mm thick copper sheet mounted in a lead frame. Openings are left in the copper sheet for the source. A plastic, *e.g.* delrin, guide ensures that foil is centered in the detector system.

An important element of the preferred implementation is the use of selected PMTs made of glass with low contamination by ^{40}K. More specifically, we disclose use of the Electron Tubes Inc. 2" or 3" PMTs preferably selected to present less than 0.1 cps background. An element of the design is decoupling the PMT base from the PMT by means of a graded shield consisting of three layers of metal with very different atomic numbers. Typically, such a graded shield consists of about 0.5" of Pb, 0.15" of Sn and about 0.1" of Cu. The use of PMT base fabricated from selected materials with low radioactive background is preferred, *e.g.*, using pure Al for the supporting frame, resistors and capacitors selected for low radioactive background, and in-free solder, *e.g.*, made of pure Sn or Sn/Pb alloy. All passive and active elements of PMT base are selected to have a very low temperature drift, and active compensation techniques to eliminate temperature dependent gain drift are disclosed.

Data acquisition in MP-densitometry is based on amplifying and shaping the signals from the PMTs coupled to each detector and building a combined energy spectrum for subsequent analysis. The counts in an appropriate energy region of interest (ROI) for the desired isotope are then integrated to determine the count rate. An important part of the preferred implementation is the use of both OR and AND modes for data acquisition and analysis. The

use of a multichannel DSO for on line background rejection is disclosed. The use of triangular shaping and software rejection of fast pulses due to signals induced by Cosmic Rays in PMT is disclosed. The particular optimization, when using $CaF_2(Eu)$ scintillators is to have a pulse rise time of < 0.75 microseconds and a fall time of about 3 microseconds.

Self-diagnostic and self-calibration for reliably matching the count rates in OR and AND modes are important, including on-line baseline restoration and pileup rejection techniques. In coincident mode the use of DSO to match the shape and temporal coincidence of pulses from two detector modules are preferred. There is a tradeoff between the need to estimate the pulse coincidence to within better than 100 nsec, diminish pileup and yet permit rejection of long duration pulse permitting rejection of dark currents from PMT. On-line software-based pulse fitting procedures overcome these conflicting requirements.

Conversion of the count rate (cpm) into the actual attenuation in the sample requires knowledge of the detection efficiency (DE) of the counter. For I^{125} the DE can be determined from the spectrum itself using the Eldridge formula. Preferably, one determines the DE for each detector separately, which allows improving the calibration and accuracy, and testing system integrity, and enhancing DE evaluation and for diagnostic purposes.

An important component of the non-radioactive background in MP-densitometer using EC sources is due to dark pulses in the photomultipliers. The shape of these pulses is different from those produced by scintillation in the detectors, making pulse-shape based discrimination possible. In calibration mode, we acquire pulse shape(s) for each event using a PC-based dual input plug-in DSO card and perform fast pulse shape analysis. This allows rejection of PMT dark pulses as well as other electromagnetic and vibrational artifacts. After pulse-shape-based rejection, the background in the system is almost flat for energies in the 15-100 keV range and is remarkably stable, independent of the activities in the vicinity of the detector.

The data acquisition hardware is mounted inside the dedicated PC controlling the MP-densitometer. The data acquisition electronics consists of a triggering circuit, amplification/attenuation modules for each detector, 3 digital timer/counters, and a dual channel 20 MHz digital storage oscilloscope (DSO) which is used both as a 2-input pulse shape/height analyzer. Furthermore, additional DSO or two ADC are used to monitor detector temperature.

The triggering circuit produces a rectangular trigger pulse whenever a pulse exceeding a preset threshold amplitude is registered in either detector. If pulses are registered

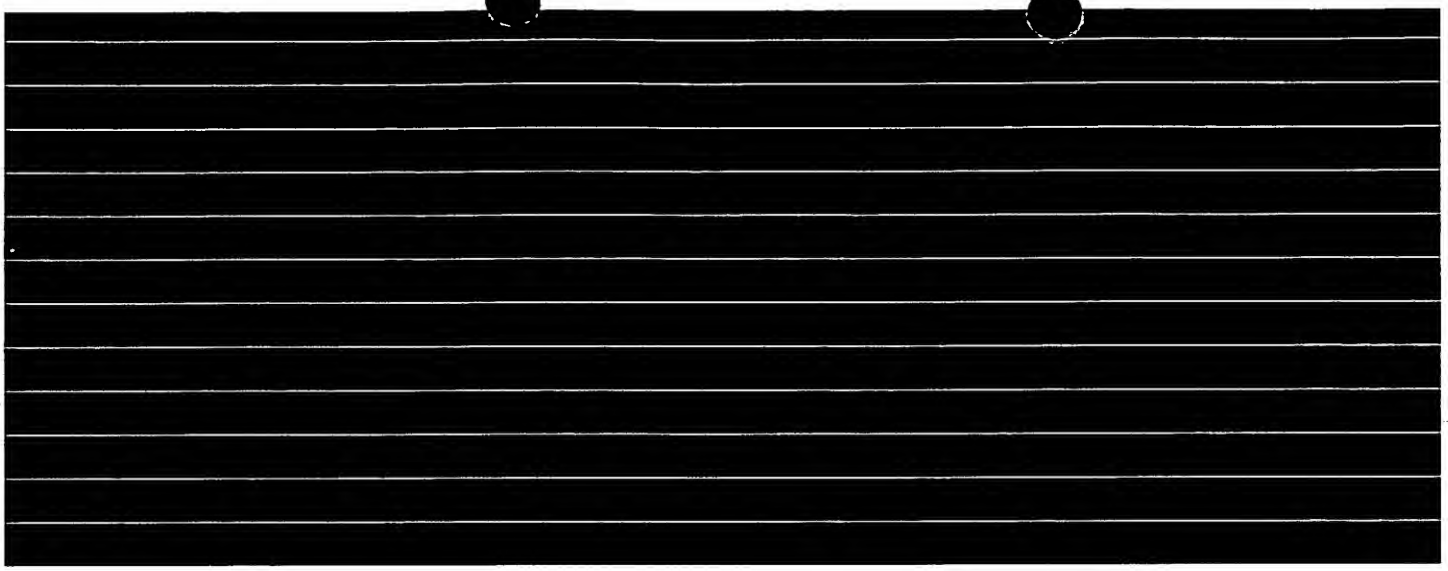
simultaneously in both detectors, a higher amplitude trigger pulse is produced. It is thus possible to count separately coincident and non-coincident events. The trigger pulse is sent to the external trigger input of the DSO or DSP. The triggering circuit is proprietary and disclosed in the following. The amplification/attenuation modules adjust the amplitudes of the pulses so that the region of interest is within the 0-1 Volt window of the DSO or DSP and that particles of the same energy produce pulses of the same amplitude in both channels.

The first of the 3 timer/counters is used as a precise acquisition time timer (counting the 2.5 kHz reference pulses). The second timer/counter counts all trigger pulses produced by the triggering circuit, while the third counts only triggers associated with coincident events. The data acquired from these counters are used to directly evaluate losses due to acquisition system dead time and thus enable the system to correctly count high activity sources.

The DSO (for example a CSLite manufactured by Gage Inc.) is capable of simultaneous sampling of two input channels with 8-bit accuracy and up to 20 MHz sampling rate and has an additional external trigger input. The data are stored in onboard memory and can be transferred to the host PC RAM by standard memory-to-memory transfer via the DSO's 8-bit access to the PC bus. The dead time is strictly non-extendable and by means of the counters described above we always correct for dead time losses. The DSO is rearmed and initialized after each acquired and processed event. The pulse traces are transferred from the DSO to the host PC memory and are analyzed for amplitude and shape by software.

Initially the DSO is set up to continuously chart the input voltages in the two channels and wait for a triggering pulse in the trigger input. When a trigger pulse is registered, the DSO is allowed to capture a predetermined number of post-trigger points and is then stopped. The relevant portion of the traces (typically 20 pre-trigger and 108 post-trigger points at 20 MHz sampling) is transferred to the host PC memory for analysis. The transfer procedure takes less than 200 microseconds per trace for a 486-DX66 computer.

The analysis begins with computation of the baseline and the pulse amplitudes in each detector. This shows whether the event occurred in detector A, detector B or both. The pulse amplitudes are adjusted for the current baselines, and if the latter are unacceptably distorted the event is rejected. Then, a number of pulse shape parameters are evaluated and compared with the ranges of acceptable values established by the software at a system setup. These include pulse widths at 1/4, 1/2 and 3/4 of peak pulse height. Fast integer-based algorithms for these computations run very efficiently on Intel processors. However, when software implementation is too slow and leads to pileup/dead-time artifacts, one may use a



are processed, they are adjusted for this dead time. The acquisition can be preset to collect data either for a given interval of time or until given statistics is acquired. The acquisition can also be terminated by the user at any time.

The software program contains a simple data file browser which allows viewing and analysis of data from single and series of measurements. The user can select a data file to be viewed through a system of menus, see the estimated density and measurement uncertainty, plot the count rates *vs.* time from beginning of measurement series and print out the data with statistical uncertainties. Both the temperature compensated and raw data can be accessed. A more intensive analysis and merging of data can be performed using a commercial spreadsheet program.

The MP-densitometer software may be coded in Borland Pascal and Assembly language (to speed up the pulse processing) and can operate under DOS using a Windows-like GUI shell, or under MS-Windows using the Delphi™ software development system (Borland International), which uses the extended Borland Pascal language. The software sets the acquisition/rejection parameters for each newly assembled detector system. This program determines the optimal trigger levels and pulse shape rejection parameters for the system and creates internal data files to store these parameters. These parameters generally do not have to be redetermined during the lifetime of the system unless a major component *(e.g.*, a PMT/base or DSO card) is replaced.

The embodiments illustrated and discussed in this specification are intended only to teach those skilled in the art the best way known to the inventors to make and use the invention. Nothing in this specification should be considered as limiting the scope of the present invention. Modifications and variations of the above-described embodiments of the invention

are possible without departing from the invention, as appreciated by those skilled in the art in light of the above teachings. It is therefore to be understood that, within the scope of the claims and their equivalents, the invention may be practiced otherwise than as specifically described.

5

CLAIMS

What is claimed is:

1. A method for determining the density of a sample comprising:

(a) placing a radioactive source comprising a radioisotope capable of concurrently emitting at least two photons so that emitted photons pass through the sample,

(b) detecting at least two photons concurrently emitted by the radioisotope, as signals in at least two detectors, and

(c) measuring the attenuation in the sample of at least one of the photons concurrently emitted by the radioisotope.

2. The method of claim 1 wherein the radioisotope is selected from the group consisting of long life positron-gamma radioemitters listed in Table 1, long life nuclear cascade radioemitters listed in Table 1, and long life electron capture radioemitters listed in Table 2.

3. The method of claim 1 wherein the total source activity is below 10 microCi and further comprising rejecting background in the photon detectors by (i) selecting low natural radioactive background elements; (ii) analyzing pulse height and pulse shape; (iii) using coincidence signature; and combinations.

4. The method of claim 1, further comprising shaping the signals from at least one of the detectors to have width of less than 2.0 microseconds, and using a fast coincidence circuit to diminish pile-up artifacts.

5. The method of claim 1, further comprising shaping the signal from at least one of the detectors to a width less than 0.5 microseconds and using a very fast coincidence circuit to diminish the pile-up artifacts.

6. The method of claim 1, further comprising analyzing the signals from the detectors on-line or off-line by DSO to establish the fraction of pile-ups, and using appropriate software correction to estimate the true count rate.

7. The method of claim 1, wherein at least one detector comprises a scintillator, an amplifier, and a photomultiplier tube, and further comprising methods to eliminate the temperature dependent drifts of count rate, selected from the use of elements with low temperature coefficients, temperature stabilization, temperature compensation, and combinations thereof.

8. The method of claim 7, further comprising placing the scintillator in a Dewar and stabilizing its temperature electronically with a precision of a few degrees Celsius

9. The method of claim 7, wherein temperature is stabilized by both the appropriate heater (preferably ohmic heater) and cooler (preferably Peltier element) and means to homogenize temperature by forced air flow.

10. The method of claim 7, wherein temperatures of both scintillator and amplifier are sensed with precision of at least 0.2 ∘ C by temperature to voltage converter (electronic thermometer) and wherein the equivalent voltage is then measured by a voltage sensor in a central processing unit.

11. The method of claim 7, further comprising measuring the temperature of the scintillator and the amplifier in a voltage sensor in a central processing unit by (i) sending an analog voltage signal equivalent to a temperature to an ADC or DSO voltage sensor by shielded coaxial cable, (ii) transforming the analog voltage signal equivalent to a temperature in an optocoupler, sending by fiber optics, decoding by light to voltage converter, and then measuring in the voltage sensor, or (iii) transforming the analog voltage signal equivalent to a temperature in a voltage to frequency converter, sending by shielded cable, and measuring by a frequenciometer in the central processing unit.

12. The method of claim 11, wherein the temperature measuring step is performed at least 10 times per second.

13. The method of claim 7, further comprising a statistical rejection procedure whereby a preset number of temperature measurements (N) is acquired, and temperature values that deviate from running average of temperature by more than 95% (more than two sigma) are

rejected and replaced by weighted average over the closest four measurements of temperature.

14. The method according to claim 7, wherein temperature measurements are used to compensate for count rate drifts due to temperature changes and to calculate the true count rate.

15. The method according to claim 14, wherein the temperature compensation is performed by means of a calibration curve which is a product of three polynomial curves taking into account temperature sensitivity of the scintillator, photomultiplier tube, and amplifier, respectively.

16. A method according to claim 7 wherein the temperature dependence of DSO is periodically measured with a system containing the means for signal multiplexing and the means to generate the pulses of well known and temperature independent shape.

17. A method according to claim 7 wherein a temperature gain calibration for each detector is obtained by a series of measurements in both OR and AND mode acquisition.

18. The method according to claim 1, further comprising steps to eliminate long term drifts of count rate, selected from (i) the use of external sources of X-ray and gamma-ray photons with known energies with at least two different lines, and mechanical means for removing or shielding the sources, (ii) the use of a separate measurement mode and calibration mode, and combinations thereof.

19. The method of claim 1, wherein at least three counters are used in measurement mode and at least two channels of DSO are used in calibration mode.

20. The method of claim 1, wherein calibration mode comprises the steps of measuring pulse shapes, calculating the energy spectrum, estimating pile-up and dead-time corrections, and measuring external electromagnetic interference.

21. The method of claim 1, further comprising comparing the data in the OR and AND data acquisition mode, including the acquisition of at least 100,000 events with both pulse-height and pulse shape rejection enabled followed by establishing detection efficiencies by comparing the count rate rates in the detectors A and B without the use of rejection.

22. The method of claim 1, wherein the sample is a fluid comprising a liquid, gas, or mixture, and the influence of container walls is accounted for by calibrations performed when the container is empty or filled with liquid of known density.

23. The method of claim 22, wherein the sample is either water or hydrocarbons with well known density, and the calibration source has activity lower than 0.5 microCi.

24. The method of claim 22, wherein the sample is either water or hydrocarbons with well known density, and the calibration source has activity lower than 10 microCi.

25. The method of claim 22, wherein a statistical rejection procedure is implemented, wherein a preset number of coincidence counts (N) is acquired, and count rates that deviate more than two sqrt(N) from the average value N are rejected and replaced by weighted average over closest four measurements of count rate.

26. An apparatus for measuring the density of a sample, comprising:

(a) a holder for placing a radioactive source near a sample so that photons from the source pass through the sample,

(b) at least two detectors capable of detecting at least two photons concurrently emitted by a radioisotope and generating a corresponding signal, at least one of the detectors being placed across the sample from the source and capable of measuring the attenuation and preferably the scatter of at least one of the photons concurrently emitted by the source, and

(c) a data processor for converting the signals to density measurements.

27. The apparatus of claim 26, wherein the source comprises a radioisotope of Table 1, and the detectors are operated in coincidence mode in a symmetric sandwich configuration as in drawing 2; in a modified symmetric sandwich configuration as in drawing 3; in a compensated sandwich configuration as in drawing 4; in a triangular configuration as in

drawing 8; or for thin foil or plate samples, in a compensated flat symmetric sandwich configuration as in drawing 9, in an asymmetric sandwich with separator configuration as in drawing 10, or in a modified triangular configuration as in drawing 11.

28. The apparatus of claim 26, wherein the detectors are operated in coincidence mode in an asymmetric sandwich configuration as in drawing 5; in a shifted asymmetric sandwich configuration as in drawing 6; or in a modified asymmetric sandwich configuration as in drawing 7.

29. The apparatus of claim 26, wherein the holder is a spool made of strong material with low atomic number selected from beryllium, plastic reinforced with appropriate fiber, aluminum, vanadium, titanium, and combinations.

30. The apparatus of claim 29, wherein the spool has essentially elliptic cross-section, with total cross-section surface close to the cross-sectional surface of two pipes it is joining.

31. The appparatus of claim 26, wherein the holder is placed inside a bypass made of strong material with low atomic number selected from beryllium, plastic reinforced with appropriate fiber, aluminum, vanadium, titanium and combinations.

32. The apparatus of claim 26, wherein the bypass has essentially elliptical cross-section, with total cross-section surface much smaller than the cross-section of the main pipe.

33. The apparatus of claim 26, further comprising a third, anticoincidence detector to diminish background.

34. The apparatus of claim 26, wherein at least one of the detectors comprises a scintillator composed of NaI(Tl) with a thickness of at least 2" coupled to a photomultiplier tube.

35. The apparatus of claim 26, wherein there are two detectors of substantially the same size.

36. The apparatus of claim 26, wherein at least one of the detectors has a scintillator coupled to a photomultiplier tube, the scintillator selected from BGO, BaF_2, GSO(Ce), CsF, abd CeF_3.

37. The apparatus of claim 26, wherein a radioisotope from Family 2 is used, at least one the detectors is a NaI(Tl), CaF_2(Eu) or YAP scintillator with thickness of less than 0.5" coupled to a photomultiplier tube, and background is diminished by measures selected from (i) using appropriate thick optical windows made of quartz or other ultrapure, optically transparent materials, (ii) placing an essentially cylindrical shield including at least one heavy metal component around at least one of the detectors, (iii) placing a separator around the source and close to one of the detectors, (iv) selecting photomultiplier tubes to have radioactive background of less than 0.1 cps, and combinations thereof.

38. The apparatus of claim 37, wherein the shield comprises a few millimeters of Pb/Sn/Cu, the Cu being placed closest to the scintillator and further comprising a shield between the photomultiplier tube and its base.

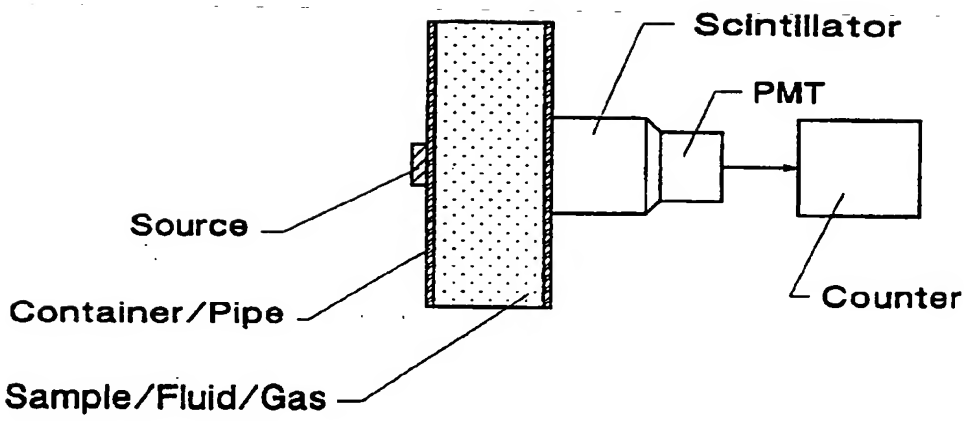

Scintillator
PMT
Source
Container/Pipe
Sample/Fluid/Gas
Counter

Figure 1

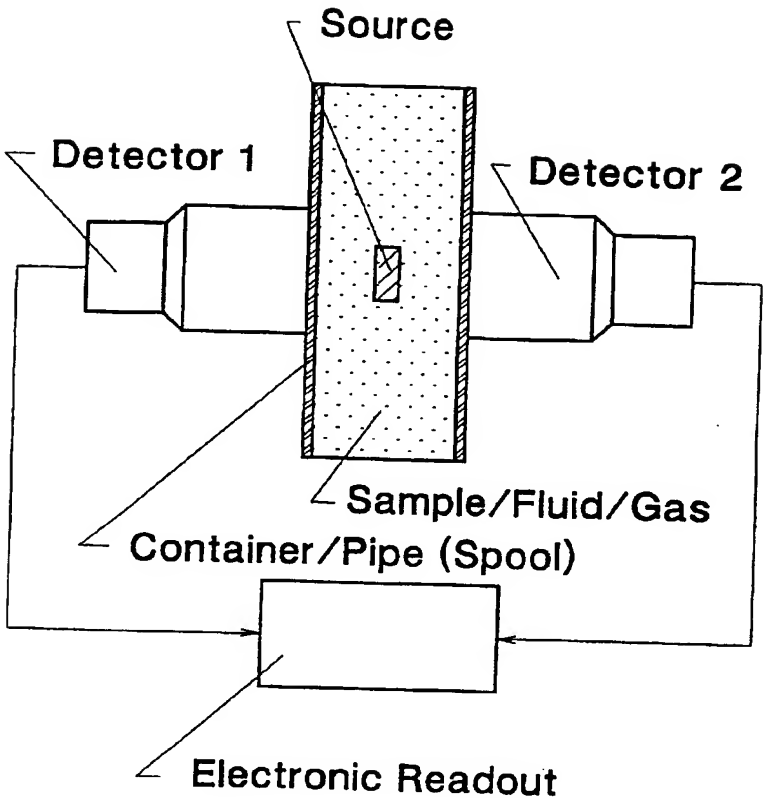

Source
Detector 1
Detector 2
Sample/Fluid/Gas
Container/Pipe (Spool)
Electronic Readout

Figure 2

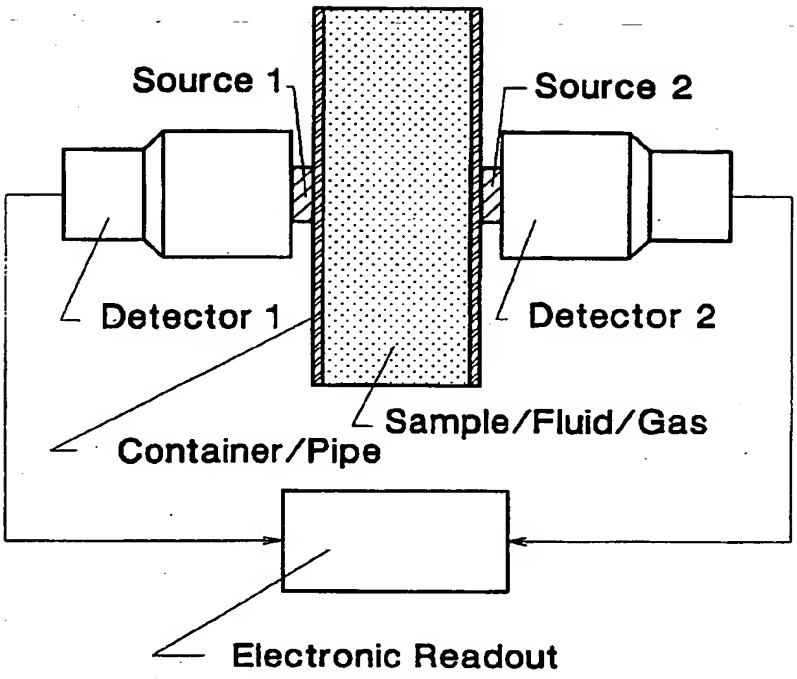

Source 1
Source 2
Detector 1
Detector 2
Sample/Fluid/Gas
Container/Pipe
Electronic Readout

Figure 3

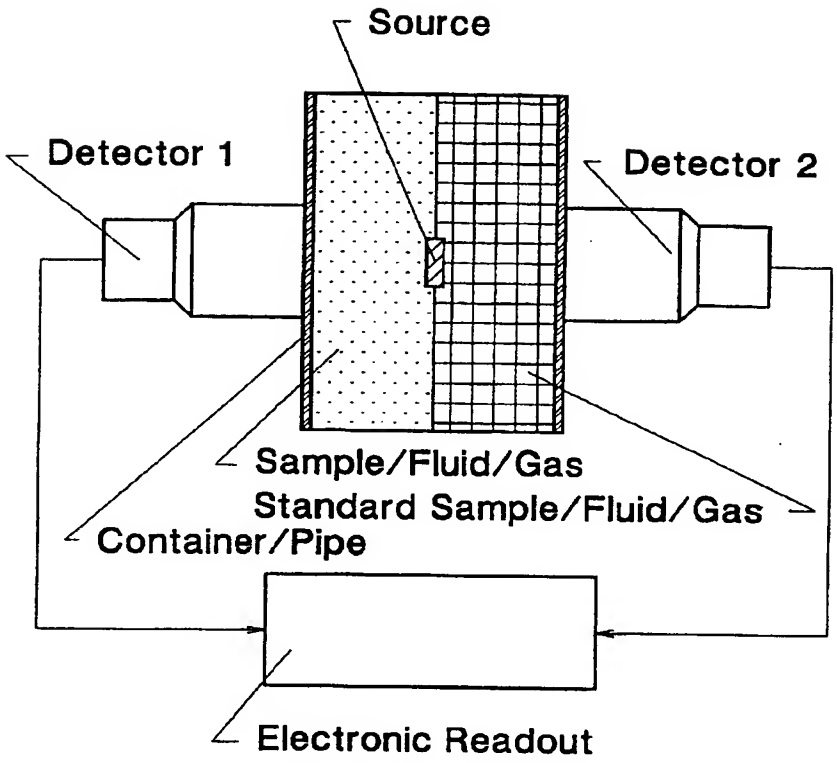

Source
Detector 1
Detector 2
Sample/Fluid/Gas
Standard Sample/Fluid/Gas
Container/Pipe
Electronic Readout

Figure 4

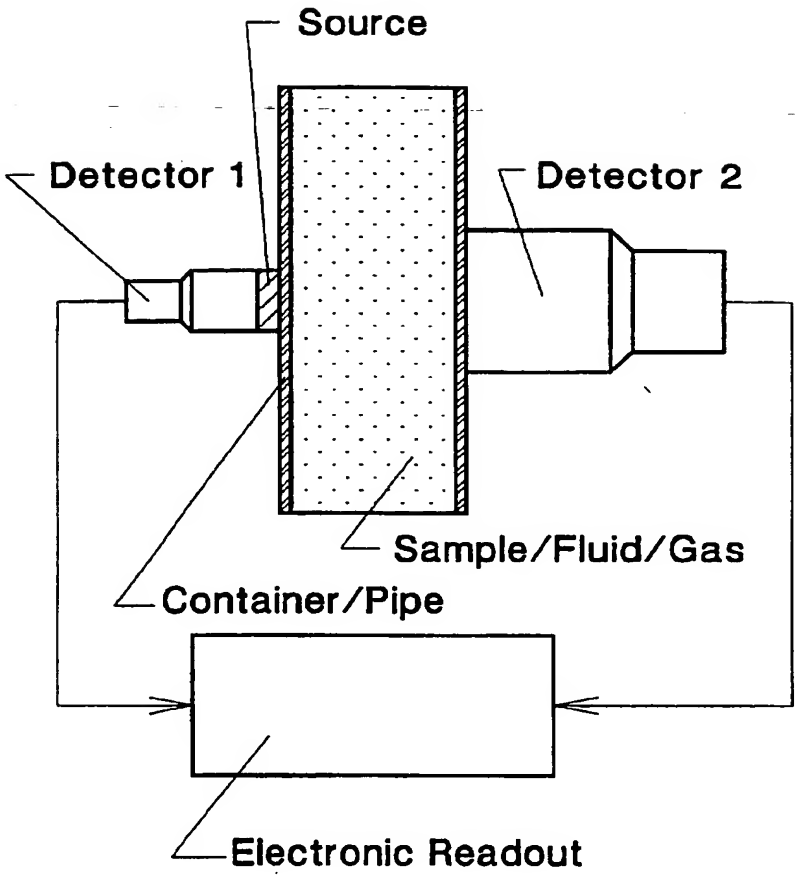

Source
Detector 1
Detector 2
Sample/Fluid/Gas
Container/Pipe
Electronic Readout

Figure 5

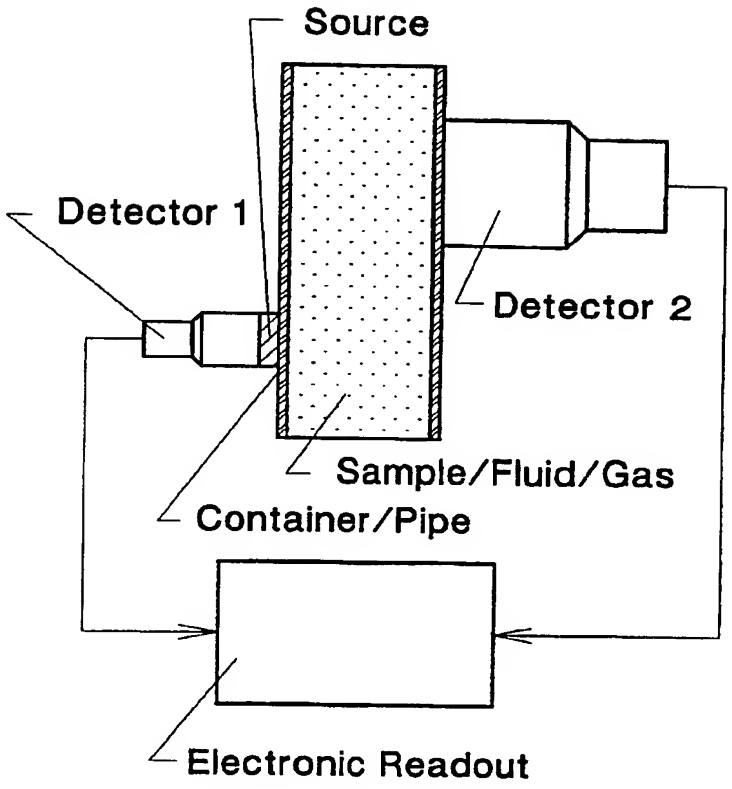

Source
Detector 1
Detector 2
Sample/Fluid/Gas
Container/Pipe
Electronic Readout

Figure 6

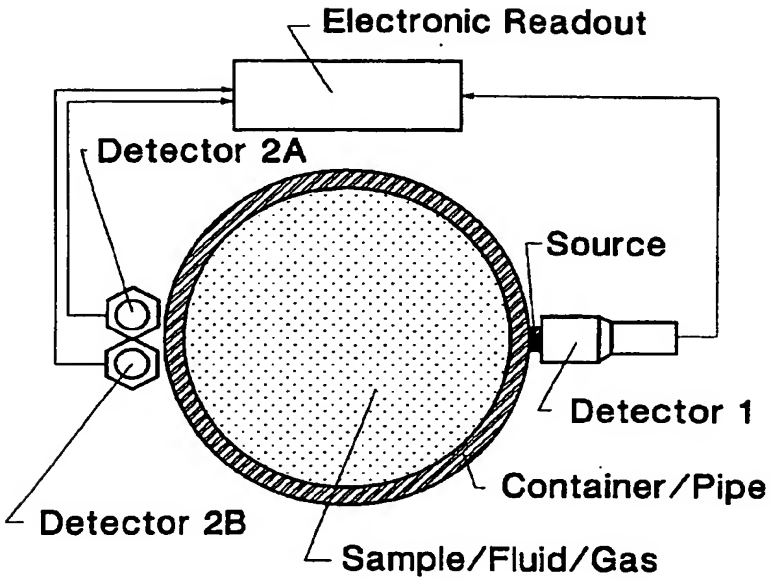

Electronic Readout
Detector 2A
Source
Detector 1
Container/Pipe
Detector 2B
Sample/Fluid/Gas

Figure 7

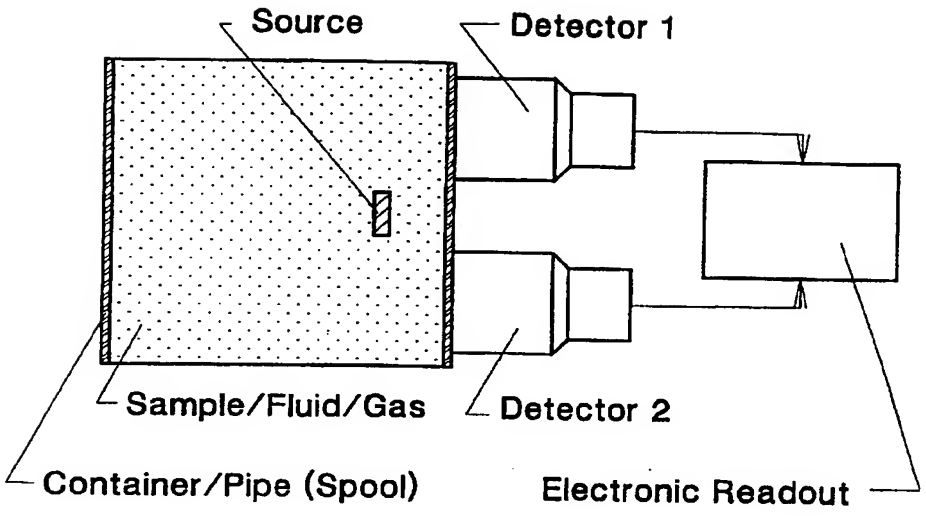

Source
Detector 1
Sample/Fluid/Gas
Detector 2
Container/Pipe (Spool)
Electronic Readout

Figure 8

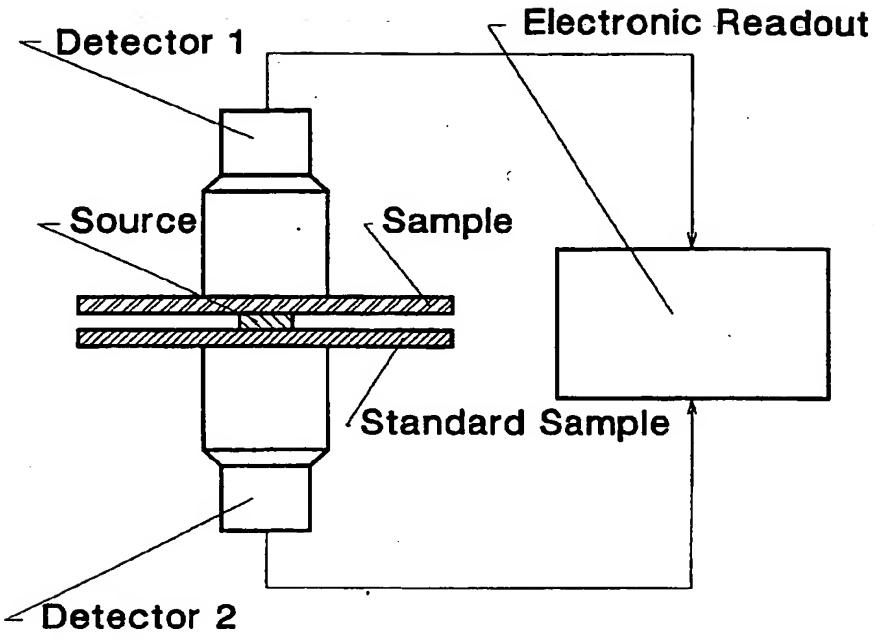

Detector 1
Electronic Readout
Source
Sample
Standard Sample
Detector 2

Figure 9

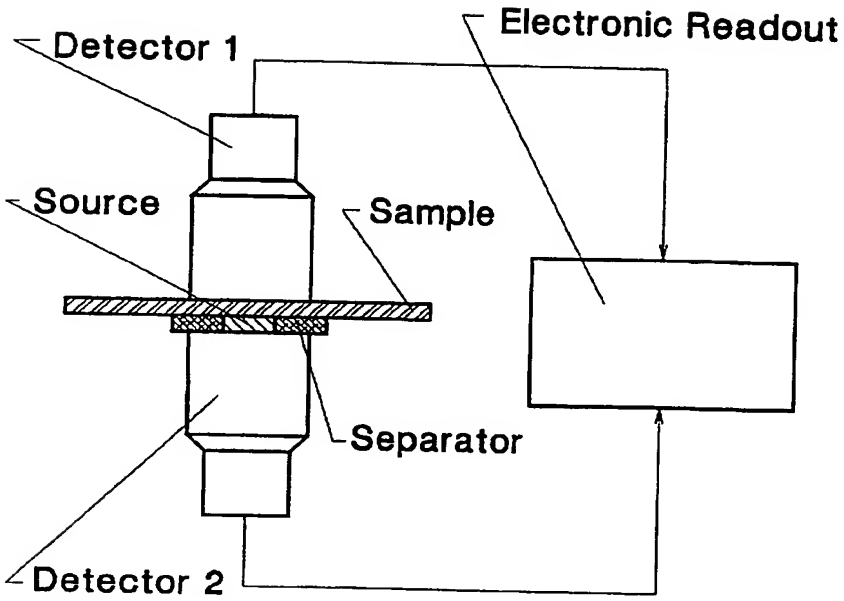

Electronic Readout
Detector 1
Source
Sample
Separator
Detector 2

Figure 10

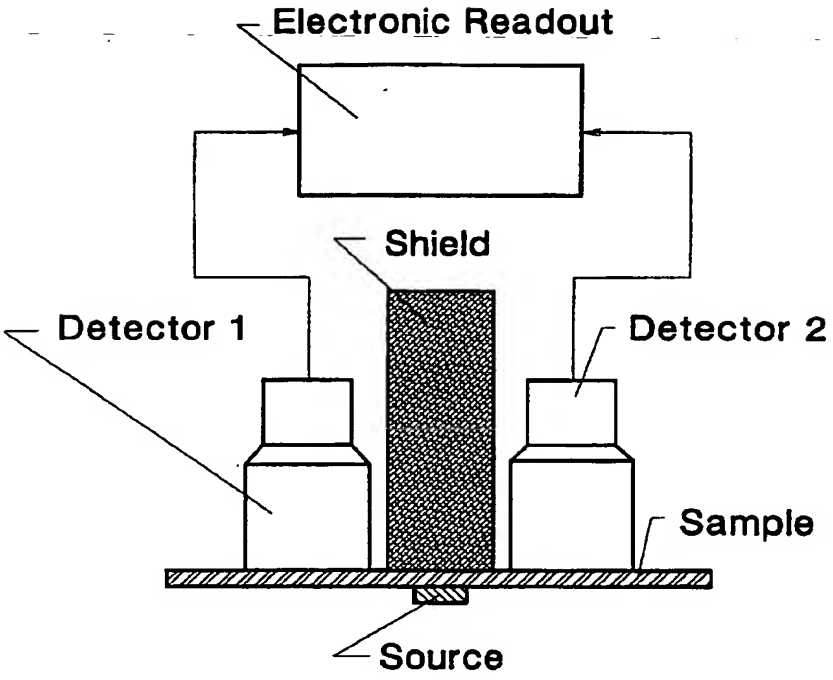

Electronic Readout
Shield
Detector 1
Detector 2
Sample
Source

Figure 11

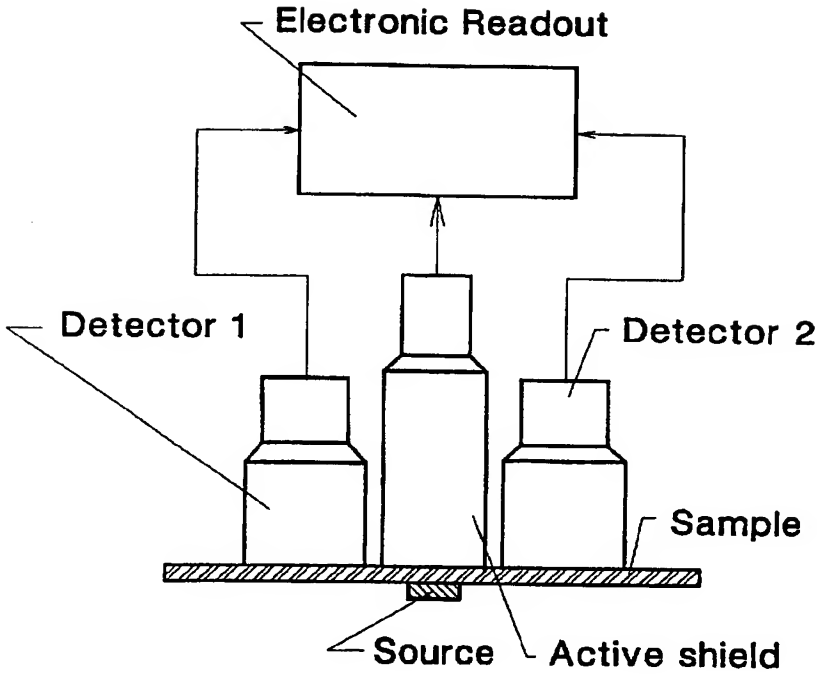

Electronic Readout
Detector 1
Detector 2
Sample
Source
Active shield

Figure 12

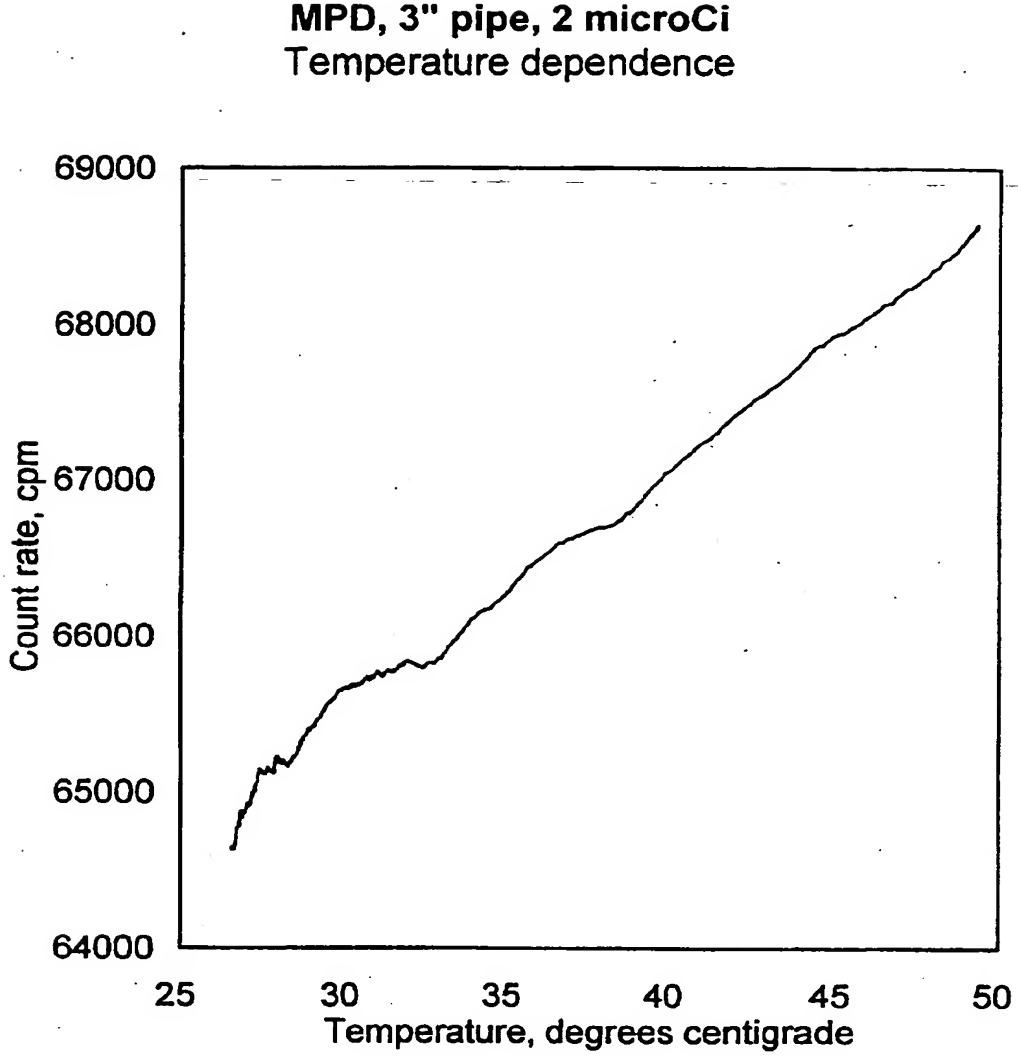

MPD, 3" pipe, 2 microCi
Temperature dependence
69000
68000
67000
66000
65000
64000
Count rate, cpm
25
30
35
40
45
50
Temperature, degrees centigrade

Figure 13

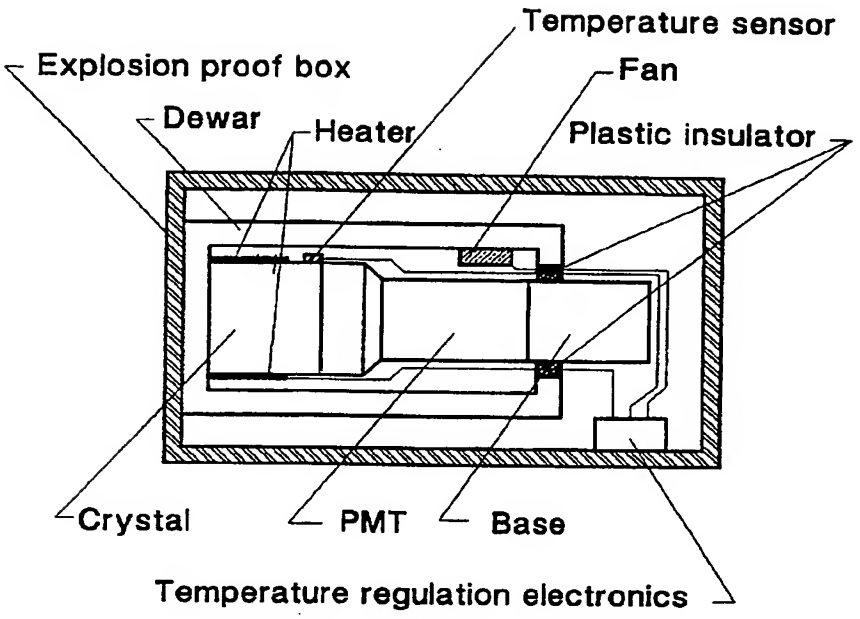

Temperature sensor
Explosion proof box
Fan
Dewar
Heater
Plastic insulator
Crystal
PMT
Base
Temperature regulation electronics

Figure 14

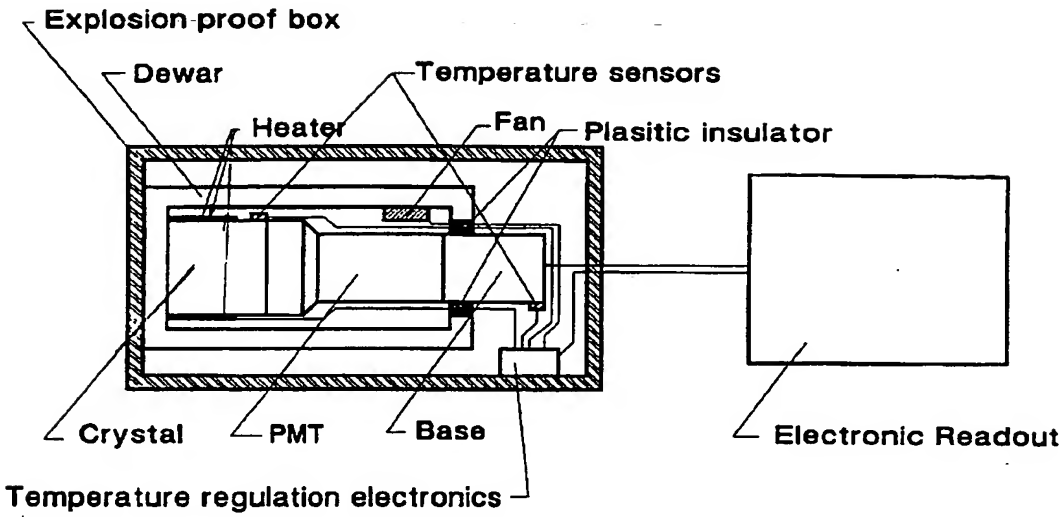

Explosion-proof box
Dewar
Temperature sensors
Heater
Fan
Plasitic insulator
Crystal
PMT
Base
Electronic Readout
Temperature regulation electronics

Figure 15

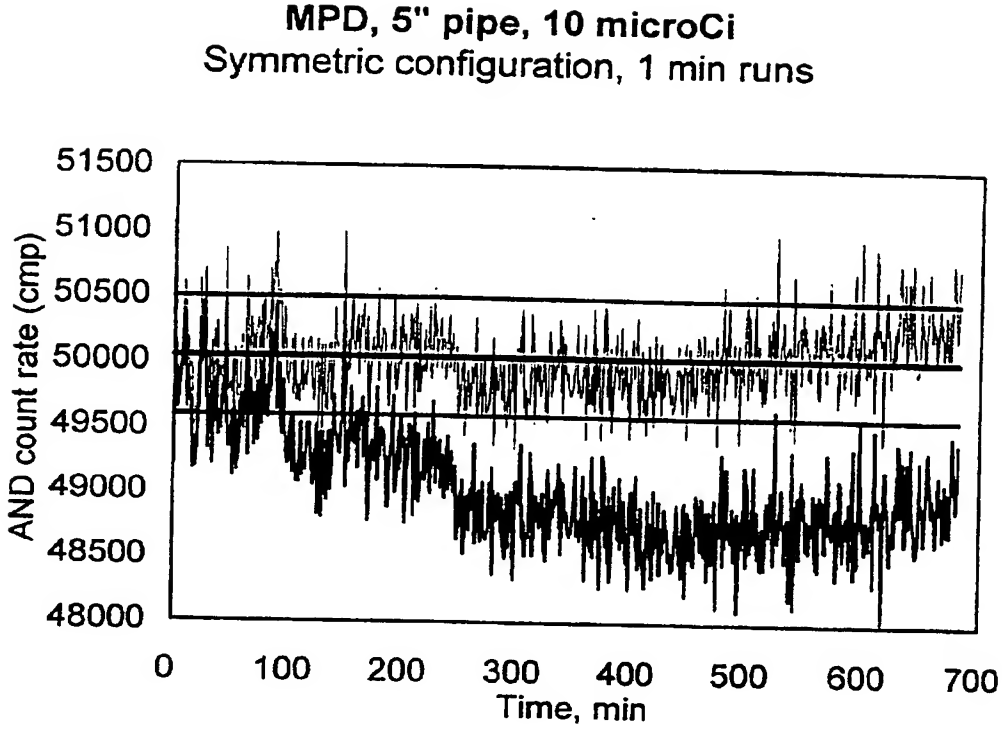

MPD, 5" pipe, 10 microCi
Symmetric configuration, 1 min runs
AND count rate (cmp)
51500
51000
50500
50000
49500
49000
48500
48000
0
100
200
300
400
500
600
700
Time, min
Count rate
Temperature corrected count rate
Mean value of corrected count rate
statistical uncertainty range

Figure 16

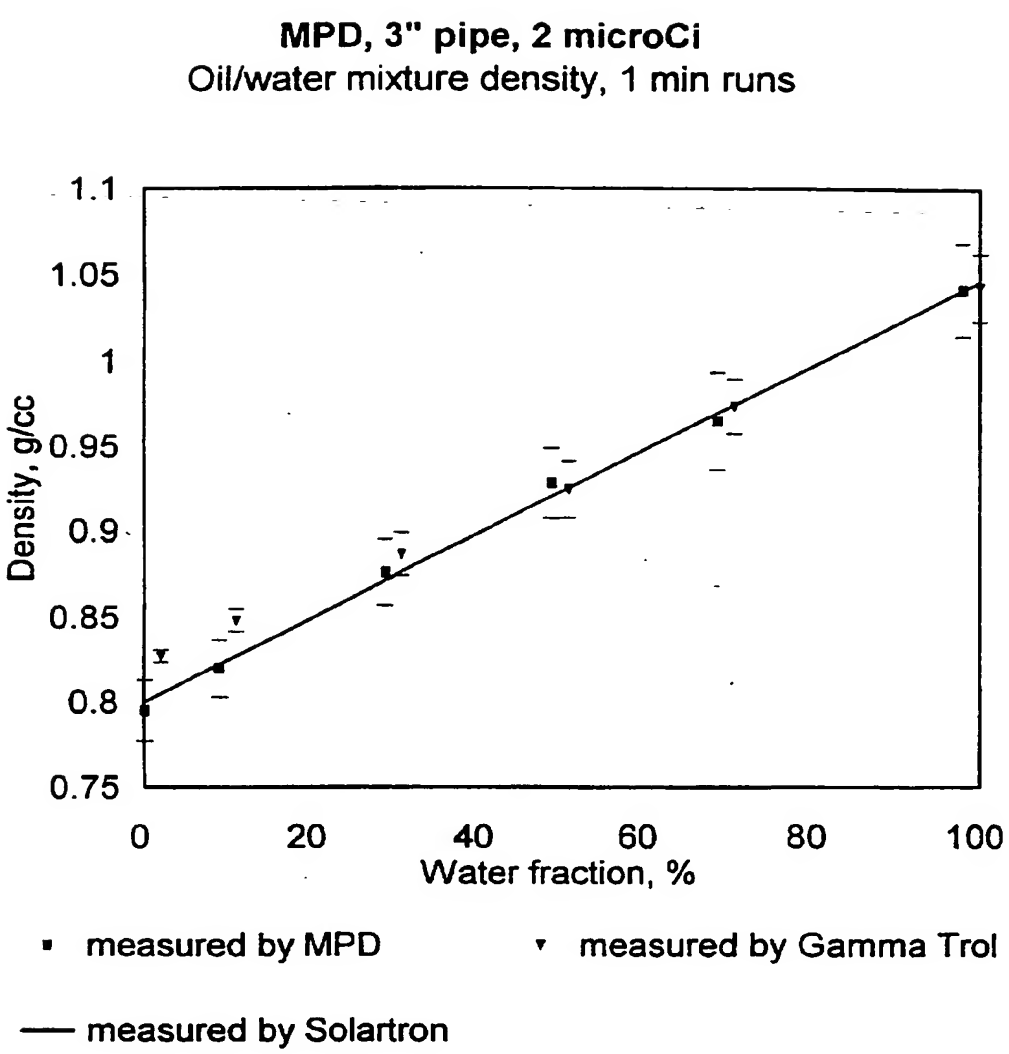

MPD, 3" pipe, 2 microCi
Oil/water mixture density, 1 min runs
Density, g/cc
1.1
1.05
1
0.95
0.9
0.85
0.8
0.75
0
20
40
60
80
100
Water fraction, %
measured by MPD
measured by Gamma Trol
measured by Solartron

Figure 17

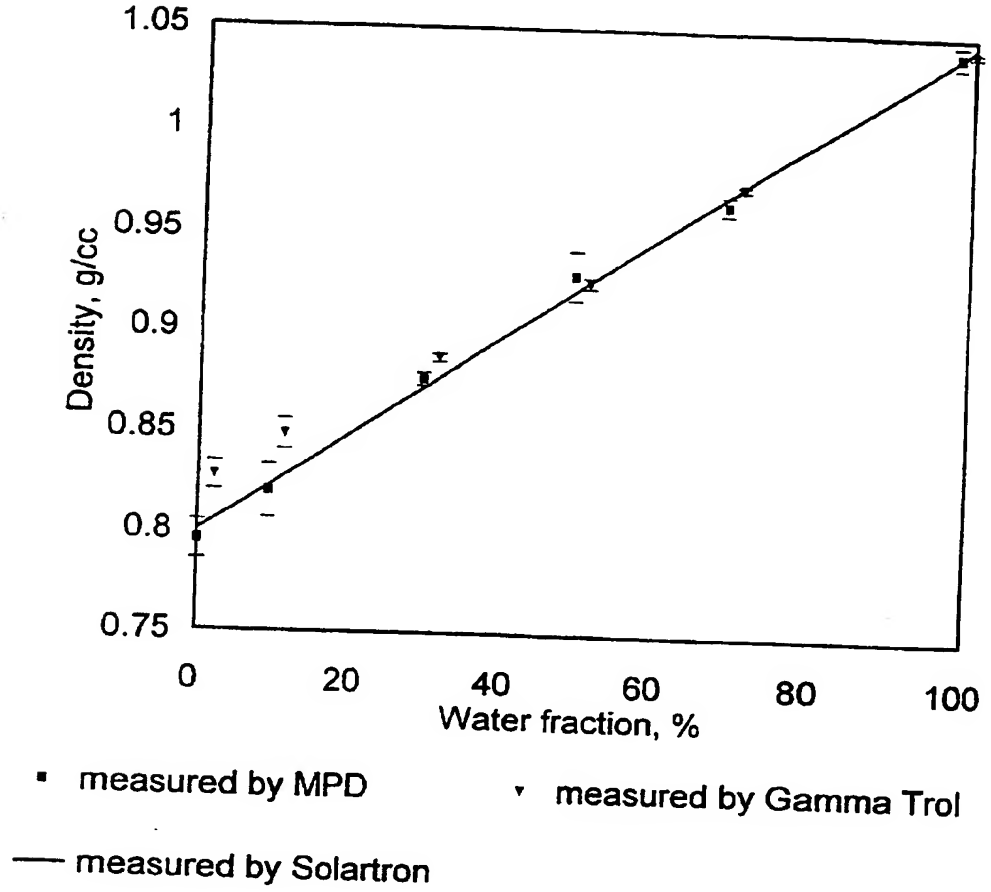

MPD, 3" pipe, 2 microCi
Oil/water mixture density, 4 min runs
1.05
1
0.95
0.9
0.85
0.8
0.75
Density, g/cc
0
20
40
60
80
100
Water fraction, %
measured by MPD
measured by Gamma Trol
measured by Solartron

Figure 18

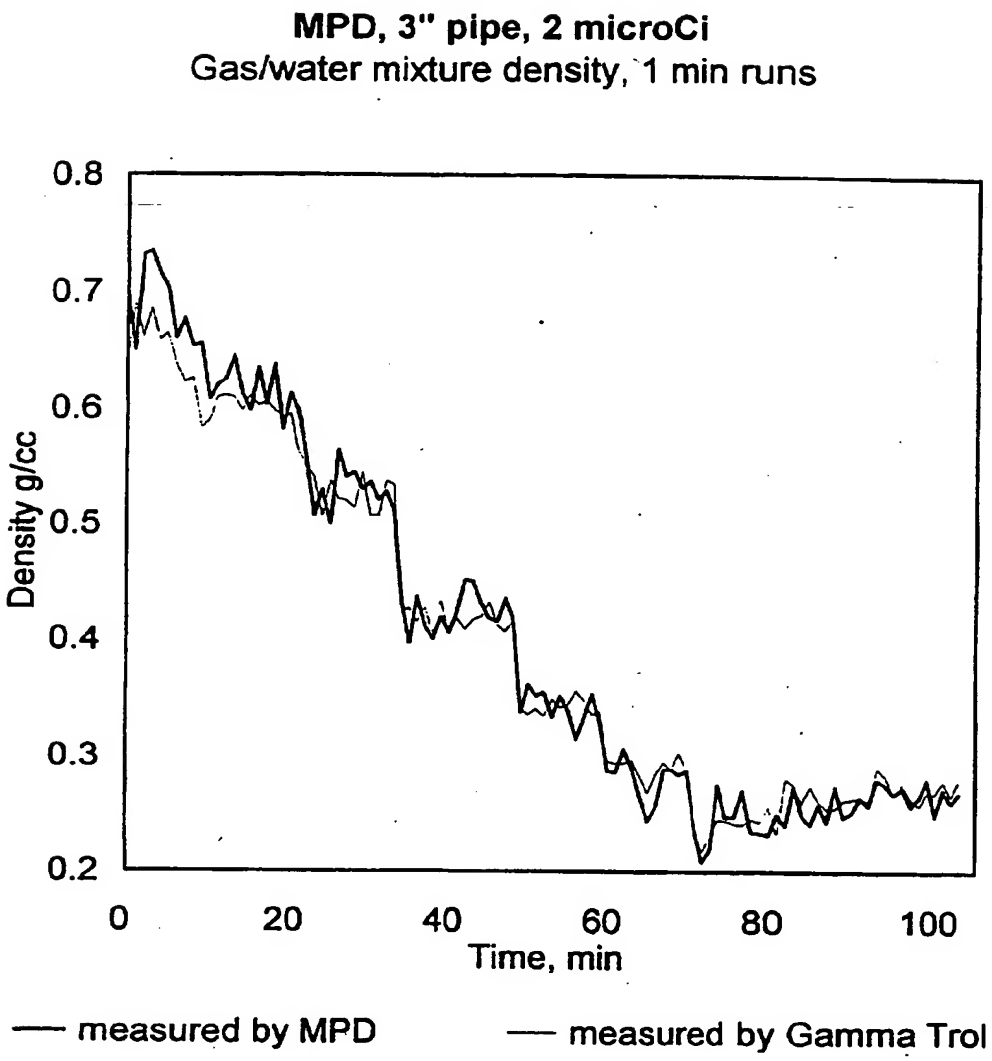

MPD, 3" pipe, 2 microCi
Gas/water mixture density, 1 min runs
Density g/cc
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0
20
40
60
80
100
Time, min
— measured by MPD
— measured by Gamma Trol

Figure 19

INTERNATIONAL SEARCH REPORT

Int. ..ational application No.
PCT/US97/02224

A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) :G01N 15/06, 29/00, 37/00, 9/00, 15/00, 23/00 G21K 7/00

US CL : 73/61.75, 64.53, 32A, 865.5, 250/308

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 73/61.75, 64.53, 32A, 865.5, 250/308

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

Please See Extra Sheet.

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 4,284,895 A (MORGAN et al) 18 August 1981, see claim 1.	1-2, 26-27
X -- Y	US 5,331,163 A (LEAHEY et al) 19 July 1994, see entire application.	1-2, 26-27 ---------- 3-25, 28-38
A, P	US 5,569,844 A (SOWERBY) 29 October 1996, see the entire patent.	1-38
A	US 4,145,917 A (BRAZHNIKOV et al) 27 March 1979, see the entire patent.	1-38
A	US 5,418,830 A (FLORENT) 23 May 1995, see the entire patent.	1-38

☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

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Date of the actual completion of the international search 19 MAY 1997	Date of mailing of the international search report 01 JUL 1997
Name and mailing address of the ISA/US Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231 Facsimile No. (703) 308-7382	Authorized officer MAX NOORI Telephone No (703) 308-5248

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INTERNATIONAL SEARCH REPORT

In ational application No.
PCT/US97/02224

B. FIELDS SEARCHED
Electronic data bases consulted (Name of data base and where practicable terms used):

APS
search terms: density measuring, radioactive, source, detecting, detector, emiting, radiating, receiving, transmitting, , attenuation